University of Zagreb

Faculty of Graphic Arts

Mia Klemenčić

THE OPTIMIZATION OF SUSTAINABLE PROCEDURES FOR THE DESIGNING AND RECYCLING OF PHARMACEUTICAL CARDBOARD PACKAGING

DOCTORAL THESIS

Zagreb, 2024.

Sveučilište u Zagrebu

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ODRŽIVI POSTUPCI ZA OPTIMIZACIJU OBLIKOVANJA I RECIKLAŽE FARMACEUTSKE KARTONSKE AMBALAŽE

DOKTORSKI RAD

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Supervisor(s)):

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Prof. Nenad Bolf, PhD

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DOKTORSKI RAD

Mentor(i):

Prof. dr. sc. Ivana Bolanča Mirković
Prof. dr. sc Nenad Bolf

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INFORMATION ABOUT MENTORS

Professor Bolanča Mirković, PhD

Professor Bolanča Mirković was born on June 7, 1976. in Zagreb. She graduated from the High School of Natural Sciences and Mathematics, enrolled in the Faculty of Natural Sciences and Mathematics of the University of Zagreb, Department of Chemistry, and graduated in 2002 with a work entitled "Determination of elements in drinking water by atomic spectroscopy and complexometry". In the same year, PhD Ivana Bolanča Mirković is employed at the Faculty of Graphics as a research assistant at the Department of Environmental Protection. In the same year, he enrolled in a postgraduate course in Graphic Engineering at the same faculty, where he obtained his master's degree on May 23, 2005, with the thesis entitled "Mechanisms of deinking in the function of the ageing of prints", and his doctorate on June 20, 2007, with the thesis entitled "Ecologically more favourable offset inks and mechanisms of deinking of prints".
As a graduate student, the Doctor of Science begins to engage in scientific work and publishes works as a result of her research. The main areas of her scientific activity are ecology, industrial ecology, the application of spectroscopic and chemometric methods in ecology, the study of methods and mechanisms of paper recycling, and the study of processes and mechanisms of natural and accelerated ageing of paints, paper and prints.

Bolanča Mirković actively participated in scientific research projects:

- "Mechanisms of deinking digital print prints" (0128003), accepted by the Ministry of Science, Education and Sports, Republic of Croatia from 2002 to 2006.
- within the scientific program accepted by the same Ministry under the title "Study of materials and processes of graphic reproduction in the function of sustainable development" (1281955), on the project "New formulations of graphic materials, characteristics of prints and environmental factors" (128-1281955-1953) from 2007 until 2010

Professor actively participated in bilateral Croatian-Slovenian projects:

- "Mechanisms of enzymatic deinking of digital prints and characteristics of wastewater" from January 2004 to December 2005
- "Mechanisms of deinking, new formulations of graphic materials and wastewater" from January 2006 to December 2007

Bolanča Mirković was the leader of the scientific project and program:

• New formulations of graphic materials, characteristics of prints and environmental factors" (128-1281955-1953) from 2011 to 2014

• Study of materials and processes of graphic reproduction in the function of sustainable development (1281955) from 2011 to 2014

Professor Bolanča Mirković published the results of her research in 14 book chapters, 31 papers in magazines, 103 papers published in proceedings of international meetings, 5 papers published in proceedings of domestic meetings and 8 professional papers.
Bolanča Mirković Ivana participated in the work of international scientific conferences as a member and president of the committee. In 2009, Bolanča Mirković was a member of the program committee of the Conference on printing, design and graphic communications, in 2011 she was a member of the expert and program committee of the International Council of Environmental Engineering Education, and in 2022 and 2023 she was a member of the Program Committee and the Organization Committee at the Printing and design conference. The professor was the president of the Program and Review Committee and the Organization Committee of the International conference on printing, design and graphic communications in 2022 and 2023, where she was also the editor of the Collection of Abstracts and Proceedings. Professor Bolanča Mirković has been the editor-in-chief of Acta graphica since October 2022.

Prof. PhD Bolanča Mirković has completed several international training courses:

• specialization at the Pulp and Paper Institute in Ljubljana for a continuous duration of 6 months (August 2005 - January 2006),

• training as part of the ERASMUS program at the Slovenská Technická Univerzita in Bratislava (STU) in the period from March 12, 2012. to March 16, 2012. (5 days),

• training as part of the ERASMUS+ program at the Instituto Politecnico de Tomar, Tomar, Portugal May 22-26, 2017. (5 days)

Doctor of Science Bolanča Mirković introduced new subjects for the doctoral study Nanotechnology and the Environment (2007) and for the undergraduate study, she introduced a program of laboratory exercises for the course Industry and the Environment. Bolanča Mirković was the immediate supervisor of 26 graduate theses and the supervisor of 47 final theses, 28 graduate theses and 1 doctorate.

Since May 20, 2019, Bolanča Mirković has been an associate member of the Croatian Academy of Technical Sciences in the Department of Graphic Engineering, and since 2022, the Deputy Secretary of the Department.

Professor Nenad Bolf, PhD

Professor Nenad Bolf was born in Zagreb, where he completed primary and secondary school. He graduated from the *Faculty of Chemical Engineering and Technology at the University of Zagreb* in 1995, obtained his Master's degree from the same faculty in 1999 and completed his doctorate at the *University of Zagreb Faculty of Chemical Engineering and Technology* in 2003 with the topic of "Adaptive Coordinated Control of Complex Processes".

From December 1995 until today, he is employed at the *Department of Measurement and Automatic Process Control* at the *Faculty of Chemical Engineering and Technology*. In the years 1995-1997 he worked as a researcher. In 1997 he was appointed junior assistant and then young assistant. In 2004 he was appointed senior assistant. In 2007 he was appointed Assistant Professor, in 2010 Associate Professor and in 2017 as Full Professor.
At the Faculty of Chemical Engineering and Technology, he teaches the courses *Measurement and Process Control*, *Matlab/Simulink* and *Process Measurement and Control* at undergraduate study and *Process Modelling and Artificial Intelligence Methods* at master study. He teaches the *Automation of Processes and Plants* course in the postgraduate *Chemistry and Chemical Engineering* programme.

He completed his education at the *Technische Universitat Wien*, *Institut fur Maschinen-und Prozessautomatisierung* in 2001 and 2002, at *Technische Universitat Graz*, *Institut fur Regelungstechnik in* 2002 and 2004; in 2005 at the *University of Maribor, Faculty of Electrical Engineering, Computer Science and Information Technology*.
His scientific field of work is the modelling, diagnosis, process control and optimisation. He is particularly interested in advanced methods of process control, process analytical technology, application of artificial intelligence, optimisation of industrial processes and thermographic diagnostics.

He is the principal investigator of the EU Structural Funds project *CrystAPC - Advanced Control of the Crystallisation Process* (2020-2023). He led one scientific project and one technological project. He was leader of a large number of cooperation projects with process industry. He is a manager of vocational and lifelong training in the process industry.

He was the national coordinator of the CEEPUS network for *Control Theory and Applications* (2000 – 2004) and the *International Study in Automatic Control* network (2005 – 2009). He is currently the national coordinator of the *Cybernetics and Modern Methods of Control network*. He was editor of the publication activities of the *Croatian Society of Chemical Engineers and Technologists* and editor-in-chief of the journal *Chemistry in Industry* (2014-2021). He was the Vice Dean for business at the Faculty of Chemical Engineering and Technology in the period from 2021 to 2023.

UDK BROJ:

Imenovano Povjerenstvo za ocjenu doktorskoga rada:

Imenovano Povjerenstvo za obranu doktorskoga rada:

Mentori:

prof. dr. sc. Ivana Bolanča Mirković, Sveučilište u Zagrebu Grafički fakultet

prof. dr. sc. Nenad Bolf, Sveučilište u Zagrebu Fakultet kemijskog inženjerstva i tehnologije

Datum obrane doktorskoga rada:

Mjesto obrane doktorskoga rada:

Povjerenstvo za obranu doktorskoga rada donijelo je sljedeću odluku:

ACKNOWLEDGEMENTS

"A good mentor teaches you how to think, not what to think."

I express gratitude to my mentor, professor Ivana Bolanča Mirković, PhD for her valuable advice, guidance, and encouragement throughout writing process of my doctoral thesis. I received support, positivity, and laugs every step way. I could not have done this without her. I must also thank professor Nenad Bolf, PhD.

I am extremely thankful to my entire family for support, encouragement, and love.

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ABSTRACT

Paper is a flat material made of plant cellulosic fibers, recycled fibers, non-fibrous components (minerals and additives) and water. It is a flexible, renewable and biodegradable raw material. Paper and paperboard are widely used as packaging materials in the global packaging market. Due to its poor barrier properties plain paper is not suitable for food packaging. To overcome this problem, paper is usually coated with other materials which often improves the barrier properties. Packaging is known as a protective outside layer of a product, and because of their similar properties food and pharmaceutical packaging undergo the same regulations. In this research three methods of removing impurities from paper pulp obtained from pharmaceutical packaging are compared and their efficiencies are determined to gain deeper insight into the recycling process. The research also studied the influence of individual phases of production of packaging on the properties and characteristics of recycled paper. The obtained optical results of the characteristics of recycled laboratory sheets were compared in order to determine the impact of each stage of box production on the quality of the paper pulp. Also, the mass fraction of metals in the different phases of the deinking process are determined. As a result, the mass fractions of metals in cellulose pulp were divided into four groups according to the mass fraction's increasing value and the metals' increasing electro-negativity. The quantities of metals were analysed using Inductively Coupled Mass Spectrometry (ICP-MS). The results of the study show that the recycling process removes certain heavy metals very well, which indicates the good recycling potential of pharmaceutical cardboard samples. The acquired knowledge is applied in the design phase of a more sustainable product to optimize the cellulose pulp's quality and design high-quality packaging products. An improved design of pharmaceutical packaging is proposed. The research focus is on the redesign of the packaging product and its impact on packaging performance. Designing for recycling will contribute to an increase in the quality of the obtained paper mass, which is directly related to an increase in the productivity of recycling and the sustainability of the packaging production process. Therefore, future research will be planned and encouraged based on the guidance gained from this investigation.

KEYWORDS: pharmaceutical packaging, packaging production phases, circular economy, flotation deinking, properties of recycled fibres, sustainable design

PROŠIRENI SAŽETAK

Papir je plošni materijal čija se građa najčešće sastoji od biljnih celuloznih vlakana, recikliranih vlakana i nevlaknastih komponenti (minerala i aditiva). Papir je fleksibilna, obnovljiva i biorazgradiva sirovina zbog čega ima široku primjenu na tržištu ambalaže. Većina današnjeg papira proizvodi se od pulpe crnogoričnih stabala (smreka i bor) koja rastu u sjevernom klimatskom umjerenom pojasu Sjeverne Amerike i Europe. Papir i karton uvelike se koriste kao materijali za pakiranje na globalnom tržištu ambalaže. Najviše se koriste nakon plastike i čine oko 37% cjelokupne ambalaže za hranu. Potrošači su skloni jesti hranu u papirnoj ili kartonskoj ambalaži.

Osnovna svojstva papira su fizička, kemijska, optička, te funkcionalna. Zbog loših površinskih svojstava i smanjene otpornosti prema vlazi, obični papir nije prikladan za pakiranje hrane. Stoga se papir često oblaže drugim materijalima koji pojačavaju njegovu otportnost. Materijali koji se najčešće koriste za pojačavanje otpornosti ambalaže izrađene od papira i kartona su na bazi plastike, stakla i metala. Uporaba ovih materijala otežavaju proces recikliranja, te samim time povećavaju ukupne troškove recikliranja. Sve dodatne komponente koje se koriste u izradi i sastavljanju ambalaže mogu biti izvor ljepljivih nečistoća (npr. smole iz drva, veziva za premaze, veziva za tiskarske boje, premazi, impregnacije, ljepila). Tako se primjerice aditivi i punila koriste kako bi se osigurala posebna uporabna svojstva papira, dok premazi i lakovi poboljšavaju površinska svojstva papira.

Primarna funkcija ambalaže jest da zaštiti i sigurno transportira, skladišti, i dostavio proizvod do krajnjeg potrošača. Ambalaža stoga štiti proizvod od potencijalnih fizičkih, mehaničkih, kemijskih, mikrobioloških i klimatskih utjecaja. Zbog sličnih svojstava ambalaža za hranu i lijekove podliježe istim pravilima i propisima. Osim što pruža fizičku zaštitu proizvoda, može produžiti rok trajanja, te održati i/ili povećati kvalitetu i sigurnost hrane. Bitno je naglasiti da ambalaža može izazvati emocionalne reakcije te na taj način motivirati potrošače za kupovinu.

Papir kao i materijali na bazi papira jedni su od najstarijih i najčešće korištenih materijala koji dolaze u direktni doticaj s hranom, a problem nastaje kada iz takve ambalaže toksini migriraju u hranu. Studije o materijalima na bazi papira za ambalažu hrane objavljene u posljednja dva desetljeća identificirale su različite tvari koje mogu biti prisutne u recikliranoj pulpi . Takav prijenos kemijskih spojeva između hrane i pakiranja naziva se "migracija". Tijekom godina

objavljene su studije koje se bave migracijom toksičnih tvari iz papira, kartona i plastike u hranu. Više od 10 000 kemikalija namjerno se koristi u proizvodnji materijala koji dolaze u dodir s hranom. Te tvari ostaju u matrici materijala tijekom recikliranja papira i tako završavaju u novim proizvodima koji su izrađeni od recikliranih vlakana.
Stoga svi materijali koji dolaze u doticaj s hranom, uključujući reciklirani papir, moraju ispunjavati neke od osnovnih sigurnosnih kriterija. Zato je potrebna sveobuhvatna analizu toksičnih sastojaka koji mogu migrirati u hranu.

Teški metali su među najvažnijim zagađivačima i mogu se naći posvuda u okolišu. Posebnu zabrinutost izaziva izloženost opasnim kemijskim spojevima koji se nalaze u proizvodima svakodnevne potrošnje. Pri vrlo niskim razinama izloženosti, štetni učinci na zdravlje općenito se smatraju zanemarivima. Međutim, neke kronične bolesti povezane su s izloženošću određenim kemikalijama koje migriraju iz pakiranja hrane. Posljednjih godina bilo je nekoliko istraživanja kako bi se napravile preporuke za sigurnost kemikalija koje se koriste u ambalaži. Stoga se teški metali smatraju jednim od najvažnijih parametara u izradi papirne ambalaže. Do kemijske kontaminacije može doći u svim fazama životnog ciklusa materijala i proizvoda. Brojne toksične kemikalije poput tiskarskih boja, ftalata, površinski aktivnih tvari, sredstava za izbjeljivanje i ugljikovodika uvode se u papir tijekom proizvodnog procesa. Stoga ambalaža za hranu izrađena od papira mora proći ispitivanja na toksične kemikalije prije uporabe. Trenutno ne postoji jedinstvena tehnologija za analizu metala u materijalima ili njihove migracije, stoga se obično nekoliko mora kombinirati. Odabir metode za analizu koja će se koristiti ovisi o njihovim kemijskim svojstvima.

Tržište recikliranog papira u svijetu postoji već trideset godina, a više od 40% ukupne proizvodnje papira temelji se na korištenju sekundarnih vlakana. Proizvodnja papirnih proizvoda pridonosi negativnim klimatskim promjenama, kemijskom onečišćenju i potrošnji energije, dok recikliranje starog papira zahtijeva 28-60% manje energije od proizvodnje papira od primarnih vlakana. Ušteda energije u proizvodnji recikliranog papira iznosi 70% u usporedbi s proizvodnjom papira od djevičanskih vlakana. Recikliranjem jedne tone novinskog papira štedi se jedna tona drva, dok se recikliranjem papira za ispis ili kopiranje štedi više od dvije tone drva. Papirna industrija stvara tone različitog otpada u svim fazama proizvodnje, odlaganja i recikliranja. Zbog pozitivnog utjecaja recikliranja papira na okoliš, količina oporabljenog papira značajno se povećala tijekom posljednja dva desetljeća. Papir se može reciklirati do sedam puta jer su vlakna svaki novim mehaničkim procesom gube mehaničkih svojstava, stoga je kvaliteta recikliranog papira ponekad lošija od papira napravljenog od sirove pulpe.

U procesu recikliranja papira nakuplja se velika količina ljepljivih čestica koje se talože na površini tkanina za oblikovanje, valjaka za prešanje i drugim dijelovima stroja. U papirnom stroju ljepljive tvari uzrokuju začepljenje tkanina i hrpe, usporavaju odvod vode iz suspenzije vlakana i time smanjuju učinkovitost procesa. Te čestice mogu biti prisutne u pulpi, procesnim vodama i/ili konačnom proizvodu. Ovi spojevi dolaze iz sirovina koje se koriste u proizvodnji papira, a pojam "ljepljivo" ne podrazumijeva određeni kemijski sastav, već je izveden iz složene fizikalne i kemijske prirode mješavine organskih tvari koje su ljepljive i hidrofobne, imaju različite oblike i gušće su od vode. Kako bi se izbjegli problemi s kvalitetom gotovih proizvoda od papira, ljepljive čestice treba ukloniti što je više moguće u procesu recikliranja. Poznavanje svojstava ljepljivih tvari pomaže u rješavanju ovog problema. Ljepljive tvari mogu se klasificirati na temelju njihove veličine i sklonosti nakupljanju.

Glavni cilj recikliranja papira je uklanjanje tiskarske boje i drugih tvari uz zadržavanje optičkih svojstava i čvrstoće vlakana. Loša kvaliteta vlakana recikliranog papira može biti problematična u krajnjoj upotrebi papira, stoga je vrlo važno procijeniti parametre kvalitete recikliranog papira kako bi se osiguralo zadovoljstvo krajnjih korisnika. Mogućnost uklanjanja boje ovisi uglavnom o svojstvima tiskarskih boja, kao i procesu tiskanja. Učinkovitost deinking flotacije i karakteristike dobivenih celuloznih vlakana određuju se mjerenjem optičkih svojstava i korištenjem slikovne analize. Mjere se karakteristike uzoraka papira prije i poslije flotacije.

Dva su glavna aspekta o kojima ovisi kvaliteta recikliranog papira. Jedan se odnosi na kvalitetu prikupljenog papira u vidu kontaminacije, vlage i sastava. Drugi se odnosi na mogućnost recikliranja prikupljenog papira.

Za papirnu industriju važno je da se papir i karton prikupljaju odvojeno od ostalih materijala koji se mogu reciklirati. Frakcije prikupljenog papira koje se uvode u proces proizvodnje novog papira trebale bi imati što veći udio celuloznih vlakana. Istraživanja su pokazala važnost selektivnog prikupljanja papira i kartona te primjene naprednih tehnologija sortiranja i njihov utjecaj na kvalitetu recikliranog papira.

Komunalni otpad sastoji se od ostataka hrane, papira i kartona, plastike i drugih komponenti. Prema globalnoj statistici, godišnje se proizvede 2,01 milijardi tona komunalnog komunalnog otpada, s prosječnom proizvodnjom otpada po stanovniku od 0,74 kilograma. Nekontrolirana količina ambalaže dovodi do prekomjerne upotrebe materijala i energije. Procjenjuje se da će se količina otpada povećati na 3,40 milijardi tona do 2050. Stoga je upravljanje komunalnim otpadom postalo problem na globalnoj razini. Udio papira i kartona u komunalnom otpadu iznosi oko 20%, dok prehrambena ambalaža čini gotovo dvije trećine ukupnog ambalažnog otpada. Održivo gospodarenje otpadom temelji se na maksimiziranju recikliranja resursa i minimiziranju finalnog otpada. Kružno gospodarstvo temelji se na ekonomskim prednostima koje nastaju smanjenjem utjecaja na okoliš i prekomjernim iskorištavanjem resursa. Rješenja uključuju ekološki dizajn, programe za smanjenje otpada i produljenje životnog vijeka proizvoda. "Reduce, reuse and recycle" tri su osnovna načela.

Ambalaža za lijekove kao skupni pojam može se definirati kao znanost, umjetnost i tehnologija zaštite proizvoda za distribuciju, skladištenje, prodaju i upotrebu, uključujući tiskane materijale koji se koriste u završnoj obradi farmaceutskog proizvoda. Takva ambalaža prije svega mora odavati dojam preciznosti, čistoće i sigurnosti. Sektor ambalaže za lijekove nedvojbeno je sektor koji zahtijeva pozornost pri promicanju kružnog gospodarstva. Globalno gledano, količina ambalaže se povećala, te je sve veći (politički) pritisak da se smanji otpad i poveća recikliranje, međutim nedostaje istraživanj i literature na temu održive ambalaže za lijekove. Onečišćenje okoliša uzrokovano lijekovima tijekom životnog ciklusa proizvoda samo je jedno od pitanja koje bi farmaceutska industrija trebala adresirati u vođenju poslovanja. Ambalaža se neprestano razvija i daje važan doprinos uspjehu farmaceutske industrije. Lijekovi se mogu podijeliti u dva segmenta: lijekovi na recept (Rx lijekovi) i lijekovi bez recepta ili tzv. „over-the-counter medications". Ambalaža je moćan marketinški alat za promidžbu lijekova, te premda njihovu kvalitetu i sigurnost reguliraju isti zakoni i propisi, postoje neke varijacije u njihovom dizajnu i prodajnoj strategiji. Dobar dizajn može plasirati proizvod na tržište, učiniti ga privlačnim korisniku, educirati ga o njegovim prednostima te pružiti upute o pravilnom korištenju.

Održiva ambalaža trebala bi biti izrađena po mogućnosti od prirodnih i obnovljivih materijala. Na kraju životnog vijeka trebala bi se moći reciklirati, ponovno koristiti ili razgraditi. Izrada ambalaže je znanstvena interdisciplinarna kategorija koja se bavi proizvodom od koncepta do kraja njegovog životnog vijeka ili uporabe koja obuhvaća područja ekonomije, ekologije i društva. Kako bi se smanjio ukupni utjecaj ambalaže na okoliš, istaknut je trend razvoja održive ambalaže. Dobro dizajnirana ambalaža može dovesti do značajnih ekoloških prednosti.

U ovom istraživanju uspoređuju se tri metode uklanjanja nečistoća iz papirne pulpe, te se utvrđuje njihova učinkovitost kako bi se dobio dublji uvid u proces recikliranja. Sva tri primijenjena postupka uspješno su odvojila čestice nečistoće iz papirne pulpe i otisnute laminirane ambalaže na održiviji način, bez upotrebe kemijskih sredstava. Ovime se također dobio uvid u utjecaj svake faze proizvodnje ambalaže za farmaceutske proizvode, odnosno na svojstva i karakteristike recikliranog papira. Uspoređeni su dobiveni rezultati optičkih karakteristika recikliranih laboratorijskih listova dobivenih iz uzoraka laminiranog i nelaminiranog kartona kako bi se utvrdio utjecaj svake faze proizvodnje kutije na kvalitetu papirne pulpe.

U drugom dijelu istraživanja utvrđuje se maseni udio metala u različitim fazama procesa deinkinga kako bi se optimizirala kvaliteta celulozne pulpe i dizajnirao visokokvalitetni ambalažni proizvod. Kao rezultat toga, maseni udjeli metala u celuloznoj pulpi podijeljeni su u četiri skupine prema rastućoj vrijednosti masenog udjela, kao i rastućoj elektronegativnosti metala. Količine metala analizirane su pomoću spektrometrije mase uz induktivno spregnutu plazmu (ICP-MS). Na odvajanje metala iz celulozne pulpe utječe prisutnost ljepila i elektronegativnost metala. Rezultati istraživanja pokazuju da proces recikliranja vrlo dobro uklanja određene teške metale, što ukazuje na dobar potencijal recikliranja uzoraka farmaceutske ambalaže.

Stečeno znanje se zatim primjenjuje u fazi projektiranja održivijeg proizvoda, gdje se laminirani materijali koriste samo parcijalno. Fokus istraživanja je na redizajnu ambalaže i njegovom utjecaju na učinkovitost pakiranja. Održiva ambalaža pridonijeti će povećanju kvalitete dobivene papirne pulpe, što je u izravnoj vezi s povećanjem produktivnosti recikliranja i održivosti procesa proizvodnje ambalaže. Stoga se planiraju daljnja istraživanja na temelju smjernica dobivenih ovim istraživanjem.

KLJUČNE RIJEČI: farmaceutska ambalaža, faze proizvodnje ambalaže, kružna ekonomija, flotacijski deinking, svojstva recikliranih vlakanaca, održivi dizajn

LIST OF PUBLICATIONS

This thesis is a summary of the following papers, which are referred to in the text by their roman numerals. The papers are appended at the end of the thesis.

I. Klemenčić, M.; Bolanča Mirković, I.; Bolf, N.; Markić, M. Determination of the Mass Fractions of the Heavy Metals in the Recycled Cellulose Pulp. *Polymers* **2024**, 16, 934. doi.org/10.3390/polym16070934

II. Klemenčić, M.; Bolanča Mirković, I.; Bolf, N. The Influence of the Production Stages of Cardboard Pharmaceutical Packaging on the Circular Economy. *Sustainability* **2023**, 15, 16882. doi.org/10.3390/su152416882

III. Klemenčić, M.; Bolanča Mirković, I.; Bolf, N. The efficiency of the separation of impurities from cellulose pulp obtained from pharmaceutical laminated cardboard packaging // *Tehnički vjesnik*: znanstveno-stručni časopis tehničkih fakulteta Sveučilišta u Osijeku, 29 (**2022**), 4; 1295-1300. doi: 10.17559/tv-20210831164929

1. INTRODUCTION

The circular economy in the production of paper and cardboard plays a key role in reducing the industry's impact on the environment and promoting more sustainable resource management [1]. The basic principle of the circular economy in the production of paper and cardboard is to maximize the reuse and recycling of materials and products as much as possible. The mentioned method of management contributes to the reduction of the need for the use of new raw materials and the generation of waste [2]. Traditional paper and cardboard production relies significantly on wood as a primary raw material. By introducing circular practices, such as increased use of recycled paper, the need to cut down new forests is reduced economy [3]. This contributes to the conservation of forest resources and biodiversity. Responsible forest management requires sustainable management of forest resources and the planting of new seedlings. The use of sustainable raw materials, which includes wood, which is greater than the natural regeneration capacity, has a negative impact on the environment [4].

Production of paper from recycled materials generally consumes less energy than production from virgin materials. By using less energy, especially from fossil sources or non-renewable sources, the emission of CO_2 and other greenhouse gases is reduced [5]. The paper and cardboard industry is one of the largest users of water in the production process. Because of the aforementioned, a circular flow of water is used in paper production, which reduces the negative impact on the environment. Advanced recycling methods can further reduce water and energy consumption per unit of paper produced, further reducing the environmental footprint of the industry.

Innovations in the production of recycled paper and cardboard, which include changes in the technological production process, but also the development of paper and cardboard that is easier to recycle and the production of paper from alternative fibers that do not require deforestation (e.g. hemp, bamboo) contribute significantly to more sustainable production [6].

The mentioned innovations have a positive effect on reducing adhesive particles, increasing process efficiency and increasing the quality of raw materials.

In the scope of the doctoral thesis, the sustainability of non-laminated and laminated printed cardboard packaging for pharmaceutical products during the life cycle of the product and the quality of paper pulp, i.e. recycled paper sheets, were studied. The results of the research contributed to clarifying the factors that influence the quality of paper pulp, which can contribute to increasing the sustainability of the production process in the product design phase. Such an approach to designing a new product creates better quality paper pulp, increases the efficiency of the recycling process, reduces problems caused by the appearance of sticky particles, and reduces the waste that needs to be disposed of.

In all phases of the test, real samples were used, which further contributes to the significance of the obtained results and the relevance of the research. The samples used were of high quality, they respected the standards of human safety, norms for the packaging of pharmaceutical products and the sustainability of the production process. The results of the investigation of metals in recycled paper sheets provide insight into the analysis of health and safety risks. The contribution of sustainability will also be shown in the area of packaging product design. As research results, guidelines for the design of a more sustainable packaging product based on the research results will be given.

1.1. Objective and Hypotheses of the Research

The hypotheses of this doctoral research are as follows:

H1. By examining the production cycle of the pharmaceutical cardboard packaging of the product, it is possible to define points that particularly affect the sustainability of recycled laboratory paper sheets.

H2. Laboratory paper and foam handsheets made of paper pulp will contain different concentrations of metal, depending on the manufacturing stages of the recycling process.

H3. Knowledge gained by observing the manufacturing stages of cardboard packaging products combined with studying the quality of the recycled laboratory paper sheets can be used to optimize the design, manufacturing, and recycling of the same product.

2. PACKAGING

Packaging fulfills many purposes, the main one being to cover and protect the goods after their manufacture, to preserve their integrity during handling, transport, storage and distribution, and to ensure their integrity during use. Physical protection improving impact protection, protecting the product inside, and reducing impact damage caused by snagging, friction, vibration, and shock. It must have excellent barrier properties against the transport of various permeabilities such as moisture, gases, and fats through the packaging wall [7,8]. Packaging can act as a light shield to protect the colour of a product from deterioration, or to postpone product expiration. E.g. milk in cardboard can have a longer expiration date than the milk packed in plastic or glass bottles. Many packages today play an active role in the quality of a product by helping to maintain the desired conditions around the product [9]. But packaging is also a way of communicating with consumers. It can influence how consumers evaluate products before purchase. Furthermore, packaging can evoke emotional responses and motivate consumers to purchase a product [10].

In the global packaging market paper and paperboard are widely used as packaging materials in many different industries [11]. They are the most widely used after plastics and account for about 37% of all food packaging for it has an environmentally friendly label [12]. Over 50 % of paper and paperboard packaging is produced for the food industry, and consumers are very likely to eat food packaged in paper or cardboard [13]. It is most commonly used for food products such as milk and milk-based products, beverages, dry powders, confectionery, bakery products, etc. The two main functions of packaging in the food industry are advertising and protecting packaged food from quality deterioration due to external influences [14]. There is a variety of different products manufactured by the paper industry. In addition to the typical products already mentioned, several products have properties that make them suitable for use in architectural structures, including paperboard, corrugated cardboard, honeycomb panels and paper tubes [15]. The use of paper for food packaging dates back to the 17th century, with its use increasing in the later part of the 19th century [16]. Paper and paperboard are mainly used at the primary (in direct contact with food) and secondary level (for the transport and storage of primary packaging) [17].

2.1. Paper

Paper is a flat material made of plant cellulosic fibers, usually mechanical and/or chemical wood pulp, but also from recycled fibers, non-fibrous components (minerals and additives) and water [17]. It is cheap in production, eco-friendly and easy to recycle and re-use, and easily available material of natural origin. Paper has been part of European culture since the twelfth century, when it arrived from the Arab countries through the Iberian Peninsula. It is a material known from everyday commodities such as printing paper, books, sanitary and household products (kitchen towels, toilet paper, etc.) or packaging material. Due to the properties of flexibility, renewability and biodegradability, paper as raw material is widely used in the packaging industry [18–20]. Different paper products will have different compositions [21,22]. Although the production of printing papers has decreased in recent years due to the increasing popularity of electronic media and devices, the production of packaging has increased, and it is the packaging materials that require durability and resistance to external conditions [15]. As shown in Figure 1 cellulose is used in various areas, e.g. in biomedicine, cosmetics, agriculture, paints, drilling muds, composite materials, as an absorbent in hygiene products and in food packaging (especially as a coating for cardboard packaging) [21,23].

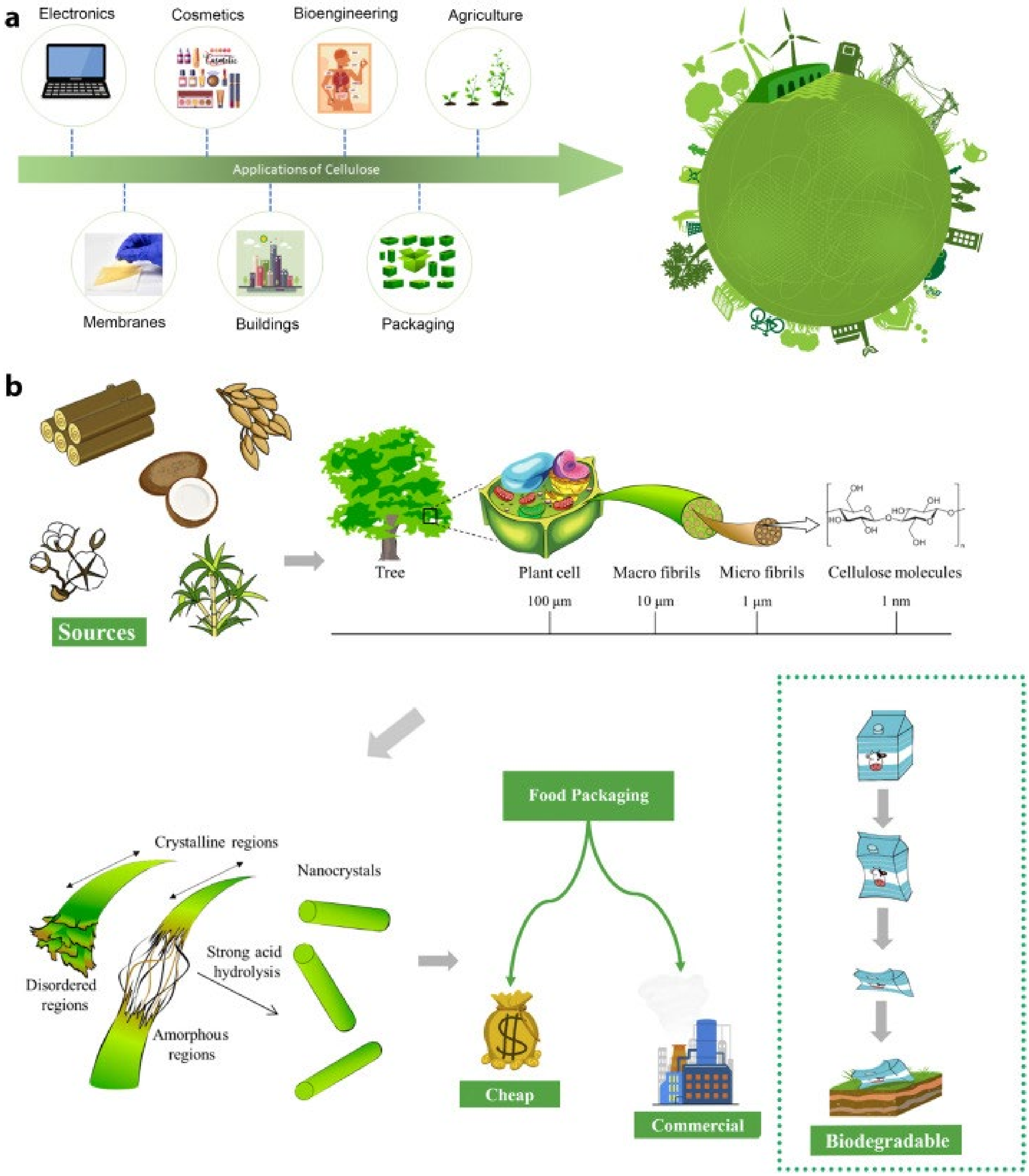

Figure 1. a) Applications of cellulose in different industries; **b)** Sources of cellulose and its applications in the food packaging industry [24]

Cellulose is the most abundant natural polymer on Earth, and the main structural fibre in the plant kingdom [25]. It is the most common natural polymer on the planet and its resources are considered almost inexhaustible. All plant matter has, on average, a cellulose concentration of roughly 33% [26]. The plant cell wall is a complex anatomical structure mainly composed of cellulose synthesized by the cellulose synthase complex. The cell wall consists of the middle lamella, primary cell wall, and plasma membrane and includes components like pectin and cross-linking glycans (*Figure 2*).

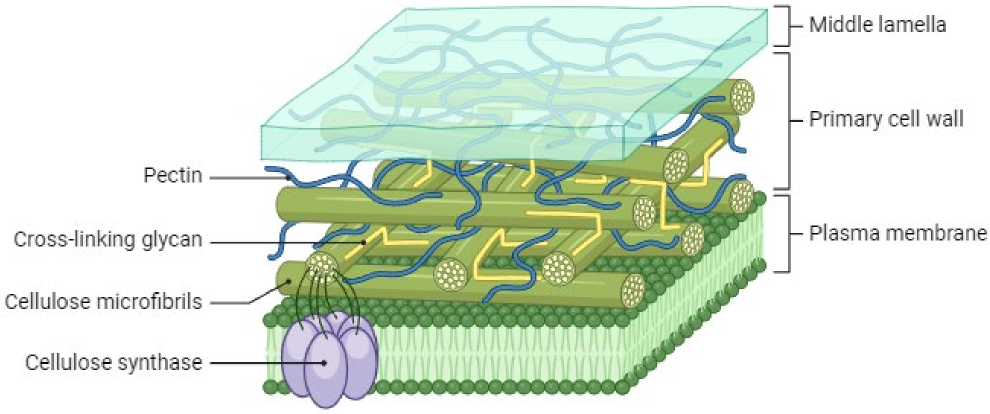

Figure 2. Plant cell wall structure (cretated by BioRender.com)

The walls of plant fibres are composed of cellulose, hemi-cellulose, lignin, extractives (pectin) and minerals. The layers are complex biocomposites made of cellulose fibril aggregates embedded in a matrix of hemicellulose and lignin. The composition of the walls of fibres can vary depending on the species and type of wood, whether it is softwood or hardwood [15]. Cellulose is a complex carbohydrate found in plant cell walls, providing rigidity and strength to plant cells. It is made up of repeating units of β-D-glucose linked together by β-1,4-glycosidic bonds. It has a linear semicrystalline structure containing a long chain of repeated D-glucose units linked by a β−1,4 glycosidic bond between D-glucopyranosyl units (*Figure 3*).

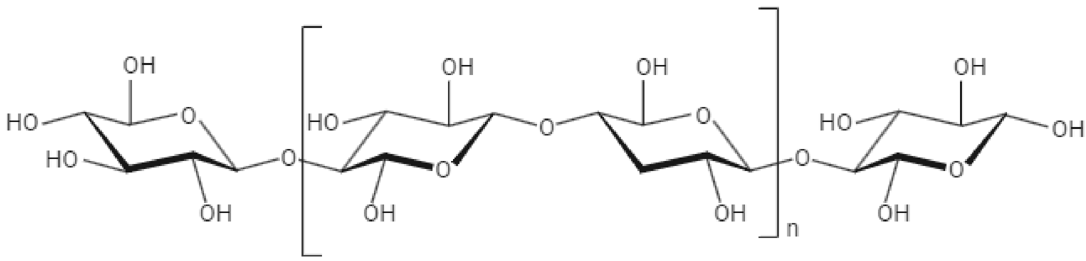

Figure 3. Chemical composition of cellulose (cretated by BioRender.com)

The key process of cellulose pulp production is the extraction of cellulose in its fibrous form [24,27]. There is a wide variety of fibre sources, as pulp can be obtained from any plant cell. The fibres can come from wood, non-timber sources or from recycled materials. The most common resource for pulp is hardwood (eucalyptus, aspen, birch, acacia, maple, oak, beech, balsam), softwood (spruce, fir, pine, cypress, hemlock, larch) and wood waste (sawmill chips and sawdust). Non-timber pulp is made from agricultural residues (sugar cane bagasse, linen and flax seeds, maize stalks, cotton stalks, wheat straw, rice straw, banana stalks), naturally growing plants (bamboo, esparto, reeds) and wood waste, Esparto, reeds, paper mulberry, sabia grass, elephant grass, switchgrass, papyrus), non-wood plants (cotton linters, cotton rags, jute, ramie, sun hemp, hemp, kenaf, palm) and textile waste. The resources for recycled paper are pre-consumer (waste generated in paper mills) and post-consumer (paper, cardboard and fibrous material collected in the packaging industry, retail, offices and households, such as printing paper, magazine waste, newsprint waste, corrugated cardboard, shredded currency waste, cotton fibres or even rhino, cow or elephant dung).

Most of the paper today is prepared from the cellulose pulp of coniferous trees (spruce and pines) whose three main components are cellulose, hemicellulose and lignin [28]. Cellulose has fiber-forming properties due to the presence of straight, long and parallel chains. It provides strength and stability to the paper structure. Hemicelluloses are a group of polysaccharides that can influence characteristics such as paper porosity, absorbency, and printability. In the process of creating pulp, most of hemicellulose is degraded, while the remaining molecules have a positive impact on the process of creating paper. They are the natural glue that facilitates the bonding offibres. Lignin is a complex polymer that is a natural binding component of wood cells that helps hold cellulose chains together. Lignin can be found both between wood cells and within cell walls. It possesses mechanical properties that make the cell rigid and provide it with a stable structure. Lignin is an undesirable ingredient in the papermaking process. Its presence causes hardening and the deterioration of the mechanical properties. Lignin is removed in the pulping process. Extractives like resin, waxes, fats, essential oils, dyes, etc. account for 5% of wood by weight. These substances may affect the properties of wood pulp. They affect resistanceto micro-organisms, but they have a corrosive effect on the production apparatus. Minerals are present in minimal quantities.

The quality of paper depends on the length and slenderness of the fibres used, as well as their resilience. The longer, slimmer and more flexible the fibres, the stronger the paper made out of them. E.g. the cells of coniferous trees are longer, more slender and moreflexible (less stiff) than the cells of deciduous trees. Therefore, they are more likely to create a strong bond during the paper-making process, which makes them more suited to the production of strong paper for packaging purposes, while the cells of deciduous trees are more suited to the creation of printing paper [15].

2.1.1. The Development of Paper Production

Prior to the invention of paper, the best writing surface was papyrus, after which paper is named. Papyrus was developed by the Egyptians and consisted of stacked reed leaves. The following row was then stacked transversely on top of the previous row. Both layers were pounded together and dried. During the pounding process, several hydrogen bonds were formed, which caused the cellulose cells to fuse. Papyrus was widely used as a writing material in Egypt and the Arabic World since 3000 BC. Paper is often associated with traditional materials and production technologies. Brought to life in the second century AD, paper has had a significant role in the history of civilisations, from the Chinese empire through the Guttenberg era up to the current digital age. It has primarily been used as an information carrier and packaging material. Although production technologies and the finish of paper have changed and improved over the years, paper has in fact remained remarkably the same through the centuries. It still has the same composition: cellulose fibres bonded in a wet environment, then pressed and dried [15].

The first paper producers on the planet were wasps and hornets, which harvested plant fibres and mixed them with their saliva to build their paper nests. However, the material today known as paper comes from the papyrus plant, which was used by the Egyptians to produce the world's first coarse writing material. Throughout the years paper is also made from cottonseed hair, flax, leaves, sunflower stalks and agricultural waste [29]. Originally, paper was used to exchange religious ideas, whether in China, the Islamic world or in Europe. Later, it was used as a general medium for written communication, i.e. for books and scrolls and the dissemination of information, as well as for official documents, contracts and money. In China, paper was used in funeral ceremonies as a representation of material goods. As an everyday commodity, paper was and still is mainly used for wrapping, but also as toilet paper, tea bags or playing cards.

2.1.2. The History of Paper Production

The invention of paper is attributed to Tsai-Lun, a Chinese minister of agriculture from the Han dynasty. Afterwards, paper became a popular medium for writing, slowly replacing silk scarves and bamboo boards as media for messages. However, paper is likely to have been invented before. Tsai-Lun was asked by the emperor to rearrange the imperial library which consisted of a large number of books made of wooden boards, which were used as a writing material at the time. In order to find handier and lighter material, Tsai-Lun began experimenting with the bark of mulberry trees, bamboo, grass, hemp, scraps of silk fibres, old fishnets and the bark of kaji trees instead of silk floss. The pulped fibres were mixed with some mucilaginous substance in a water solution. Then the material was screened, drained and dried. Although the processes, machinery and technology has changed over the centuries, paper is still made the same way it was then. When paper was invented, its production method was initially kept secret. As alightweight and relatively cheap material produced out of tree bark, rags and later fishing nets, paper replaced heavy bamboo boards and expensive silk as the preferred material on which to write. In 105 BC the experiments finally led to the first authentic papermaking process and the invention of a material consisting of plant fibres bound together to form a single sheet [30]. Before paper was introduced and adopted by other parts of the world, other materials were used as information carriers, such as bricks, lead, brass or bronze sheets, pieces of wood, the inside of tree bark, tree leaves, vellum, parchment, stone tables or papyrus.

In the beginning paper was mainly used to spread religious ideas, but also for the creation of money. Papermoney was first used in China back to 812 AD, toilet paper was first reported by travellers in 875 AD, and in 969 AD the existence of paper playing cards was reported.
The Arab world acquired knowledge of papermaking in the eighth century, after the conquest of the city of Samarkand, where several papermaking workshops had previously been established. In Islamic culture, paper was mainly used to spread the verses of the Kuran. The Arabs also improved the pulping process by inventing mechanised pulp-making involving water mills. The ingredients now no longer had to be manually beaten into a pulp. The knowledge of papermaking came to Europe with the Arab expansion on the Iberian Peninsula, where the first papermaking workshop was opened in the city of Xativa in 1144. At that time, parchment and vellum were the most common writing materials in Europe, therefore paper was not trusted as a reliable material for documents and religious books for another several

centuries. For example, the first printed books in Europe, Gutenberg's Bible, were printed between 1452 and 1455 on both vellum (45 copies) and paper (135 copies).

The European technique of paper-making differed from the techniques used in China, Japan and the Arab world. Considerable progress in pulping was made in the northern Netherlands, where wind mills were used for pulping. The second half of the 17th century was a great breakthrough in the development of papermaking because of the invention of Hollander Beater. Instead of wooden beaters moved by water mills and the fermentation process for pulp production, the machine used steel blades that cut the raw material and accelerated the pulping process. As a result, papermaking became cheaper and faster and the Netherlands soon began exporting paper.

In the 18th century, people looked for new resources needed to make the paper because old rags, hemp, and cotton were scarse. A growing demand for paper led to new breakthroughs in paper production. New raw materials for paper production were researched by people such as French physicist and naturalist René Antonie Ferchault de Réaumur, German clergyman Christian Schäffer and German inventor Friedrich Gottlob Keller. De Réaumur suggested that if wasps and hornets could produce a paper from wood, humankind should also be able to do so, while Schäffer concluded in his books that paper can be made out of any plant, and that the different characteristics of plant structure would result in different paper qualities. In 1840 Keller managed to obtain pulp from mechanically grinded wood. In the 1840s Fredrich Gottlob Keller obtained paper pulp from mechanically shredded wood. This discovery, together with the invention of Robert's machine, which produced paper in continuous stripes, triggered a revolution in the paper industry. The inventions of the eighteenth and nineteenth centuries concerning raw materials and the production of paper and cardboard resulted in a revolution in the paper industry, which in turn led to the mass and cost-effective production of paper products, and further development of the industry. And most importantly, they made paper widely available material. The history of paper development shows how paper products have been used from small objects in ancient times to large-scale structures today. Now, in the twenty-first century, the golden era of paper-making may be about to end.
Due to the advent of modern media such as tablets, computers and other digital file readers, there is less and less need for printing paper. However, the demand for packaging materials is increasing [15].

2.1.3. The Production of Paper

As mentioned afore, paper is a material of organic origin, and the most popular raw materials from which paper is made are deciduous and coniferous trees. However, paper can also be made out of other plants, such as straw, hemp, cotton, bamboo, cane and other cellulose containing materials. Using recycled paper as a source material is more and more popular.

Paper production is divided into two phases. First is the preparation of paper pulp, and second is processing the pulp in paper mills to form paper sheets. Pulp consists of small, elongated plant cells that form a compact tissue made of the raw material. The pulp used in paper production must be ground into individual fibres. Sheets of paper are produced by using the fibres' ability to form bonds with each other during a process of wetting, heating and pressing. Even though paper manufacture has seen significant technological advancement over the years, particularly in the twentieth and twenty-first centuries, paper is still a low-tech material whose mechanical properties depend on bonds between the cellulose fibres. The raw material used for the development of paper has a strong effect on the quality of the paper due to the different fibre lengths and pulp compositions. Most of today's paper is made from the pulp of coniferous trees (spruce and pine) that grow in the northern temperate zones of North America and Europe. Cellulose, hemicellulose and lignin are the three main components of the cell wall of wood. Cellulose possesses fibre-forming properties as it has straight, long and parallel fibres. Hemicelluloses are responsible for the hydration of the cellulose and the development of bonds during beating. Lignin is a natural binding component of wood cells that cannot form fibres.
To produce paper, pulp is mixed with water and poured onto the screens. This slurry consists of 99.7% water. Most of the fibres are aligned in the direction of the screen's movement (machine direction), which determines the anisotropic character of the material. The water is then drained and the pulp is pressed and dried. During this process, hydrogen bonds are formed between the cellulose fibres. The bonds between the fibres determine the mechanical properties of the material. Longer, slender, and more flexible fibres produce stronger paper. Conifers are therefore an ideal raw material for strong paper. There are various methods for obtaining paper pulp. Among them, the chemical sulphate process (Kraft) produces the cleanest pulp, which consists of around 95% cellulose and is suitable for strong paper.

During pulping, the wood fibres are separated from each other by mechanical, chemical or thermal treatments or by a combination of these processes. During pulping, lignin is dissolved to produce individual fibres that can be formed into sheets of paper during papermaking. The solution or fibres obtained after the pulping treatment are called pulp. Bleaching treatment is used to improve the whiteness of chemical and mechanical pulp.

The chromophoric groups of the lignin are responsible for the colour of the pulp and are removed during bleaching with chlorine, chlorine dioxide or hydrogen peroxide. The beating treatment increases the surface area of the fibres, which increases the water-holding capacity and creates additional binding possibilities for the fibres [17]. The refining process is similar to the beating process and serves to improve the physical properties of the finished sheet. After pulping and defibration, stock preparation (fibrous material) involves the mechanical treatment of the pulp to turn it into a sheet on the paper machine. Three different methods are mainly used in paper production: Fourdrinier machine, cylinder machine and twin-wire former. During the paper-forming process, the fibrous material (which contains about 99% water) is passed through rollers or wire mesh to remove the water and form the paper web. Final treatments include calendering, supercalendering, sizing, laminating, impregnation or saturating the developed paper, depending on the requirements of the industry or the product to be packaged [13].

Calendering is a process during which paper is run through rollers. It derives its smoothness and glossy properties from the application of pressure and heat. After the calendering treatment, the paper is referred to as machine-finished. Supercalendering and calendering are nearly identical processes. Supercalendering involves the addition of moisture and more pressure. Sizing is the process of coating paper with starch, casein, alum, etc. to improve its appearance, barrier properties and strength. Coating is a process that can be applied in the paper machine or elsewhere. Special coatings which are applied to the outer layer of the paper may, for instance, create barrier properties for special paper such as the impregnated or paraffined cardboard used in the building industry [15]. *Figure 4*. presents the paper production process, including the preparation of the pulp with additives and fillers, and the formation of paper in the paper machine.

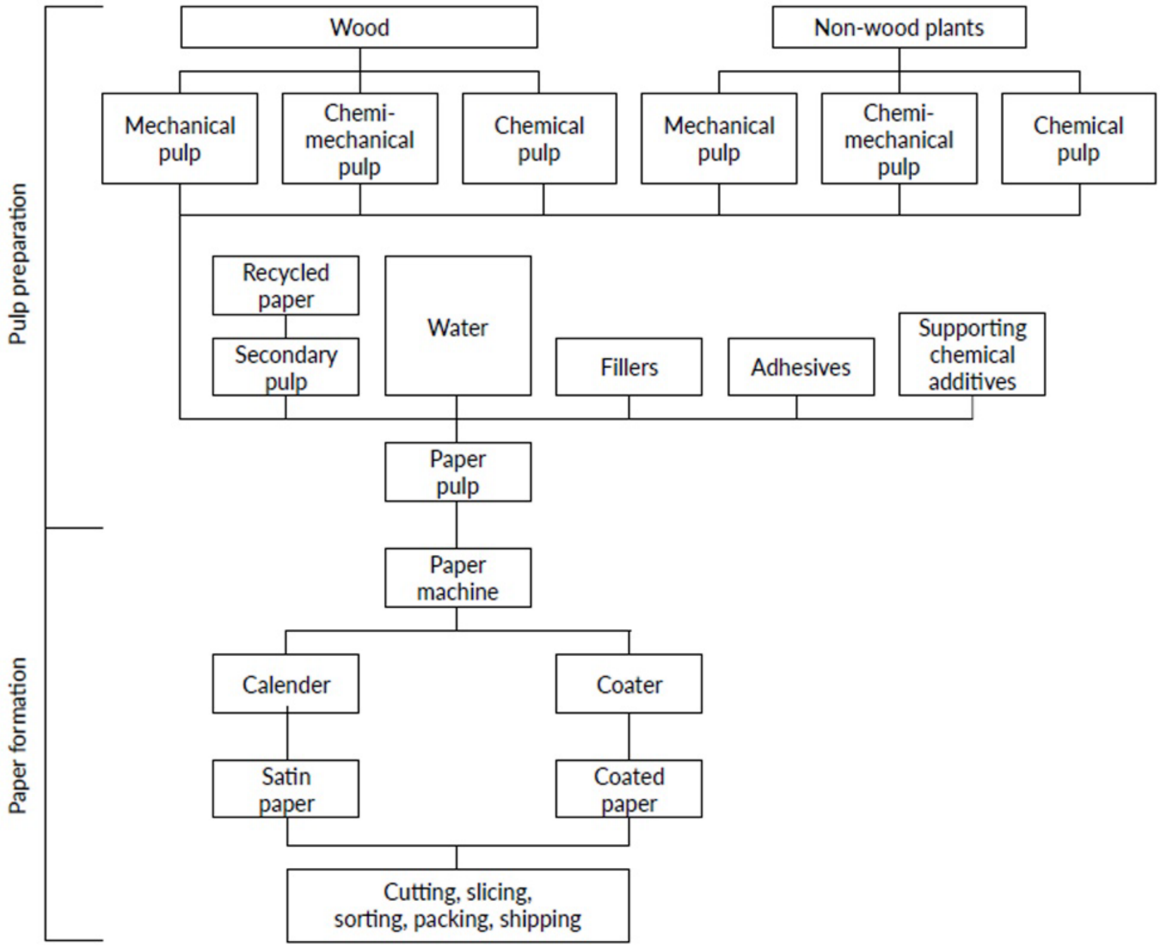

Figure 4. Paper production process [15].

2.1.4. Paper Grades

The meanings of paper and paper-based products are often confused. Usually, these materials are classified as paper (newsprint, stationery, tissues, bags, towels, napkins, etc.) or cardboard (liner board, corrugated cardboard, tubes, drums, milk cartons, recycled cardboard for shoe and cereal boxes, roofing felt, fiberboard, etc.). When talking about paper usually, paper refers to thin paper products, while cardboard refers to thicker products. Paper weighing more than 225 g/m^2 is defined by the ISO (International Organisation for Standardisation) as paperboard, board or cardboard; however, according to the definition of the Confederation of European Paper Industries (CEPI), paper is usually referred to as board if it is heavier than 220 g/m^2. Paperboard is thicker than paper and has a higher weight per unit area, although the dividing line between the two is somewhat blurred as both products have the same material composition. However, some products are labeled as paperboard even though they are produced with a lower grammages (e.g. 180 or 190 g/m^2). due to improved manufacturing processes, these lightweight materials can now be produced with similar strength properties to the older, heavier grades. For quality reasons, paperboard usually requires a combination of several layers of fibres in the wet state, it can be produced in a single ply or, more commonly, in several plies (multiply). When studying the traditional paperboard market, one can see that multiply paperboard is already produced at 160 g/m^2. Paperboard can be used for the production of food packaging such as folding cartons or paper cups. Its thickness ranges from 0.25 mm to 4 mm and its weight from 224 g/m^2 to 1650 g/m^2. To simplify the distinctions between different paper and cardboard, it can be assumed here that the difference is in grammage, where Paper is a material with a grammage lower than or equal to 225 g/m^2, and Cardboard is a material with a grammage higher than 225 g/m^2 . In multi-layered materials (i.e., corrugated board) this boundary is equal to 160 g/m^2 [31]. Therefore it can be assumed that Cardboard is a commonly used term that is associated with thick paperboard or corrugated board.

The definitions according to the NEN-ISO 4046 1-5 norm are listed below:

- Paper is a generic term for a range of materials in the form of a coherent sheet or web. In the generic sense, the term of "paper" may be used to describe both paper and board. The primary distinction between paper and board is normally based upon thickness or grammage.
- Paperboard (also board) is a generic term applied to certain types of paper frequently characterized by their relatively high rigidity.
- Carton board (also folding boxboard) is board intended for manufacture of cartons having good scoring and folding properties.
- Corrugated fiberboard is a board consisting of one or more sheets of fluted paper glued to a flat sheet of board or between several sheets [15].

2.1.5. Paper Properties

Paper has a web-like fibre network which forms during the papermaking process. After the pulp is drained, the fibres are held together by surface tension forces, which give paper its viscoelastic character. The length of a single fibre ranges from 1 to 3mm, and the width and thickness of a single fibre range from 10 to 50 μm. The number of fibres per unit area is described in terms of basis weight or grammage [g/m^2]. The thickness of paper is always specified by the grade of the paper. The thickness of paper can vary depending on the moisture content of the material. Common printing and writing paper is about 0.1mm thick. Cardboard can be 0.3 up to 4mm thick. Typical apparent density values range from 0.5 to 0.75 g/cm^3. Since cellulose density is 1.5 g/m^3, this means that 50 percent or more of most types of paper is empty space. This space is occupied by air. Apparent density is one of the most important factors affecting the mechanical, physical and electrical properties of paper. The porosity of paper is determined by its density, has a significant impact on the other properties of paper. Porosity is the ratio of pore volume to the total volume of a sheet of paper. It is akin to air permeability, which is the property of paper that allows air to flow through a sheet of paper under changing pressure conditions. Air permeability is a structure-related property of paper and is inversely related to its strength properties. It also affects paper's resistance to water and other liquid reagents [15].

The basic properties of paper are characterised by weight and density, moisture content, physical characteristics, strength properties, optical properties and other criteria.
The mechanical properties of paper can vary, even between sheets of paper made out of the same pulp because fibres in sheets are oriented randomly, so each production series may differ. The more bonds are created between cellulose fibres, the stronger the paper; therefore mechanical properties of paper are governed by fibres and the bonds between them.
The chemical properties of fibres depend on the raw material (fresh or recycled, hardwood or softwood) and pulping method used (e.g.chemical, mechanical, chemo-mechanical, etc.). In general, paper and cardboard are inhomogeneous, anisotropic, non-linear, viscoelastic-plastic and hygroscopic materials [15].

The hydrophilic behaviour of paper and its porous structure can prevent its use in several applications. Due to its poor barrier properties, low heat sealability and strength, plain paper is not suitable for food packaging [32,33]. To overcome this problem, paper is usually coated in combination with other materials such as plastic or aluminium, which often improve the barrier properties at the expense of its environmentally friendly and biodegradable properties [34]. At the end of the 19th century, several technologies were developed to provide paper used for food packaging with good barriers (e.g. greaseproof and glassine paper, based on intensive mechanical treatment of the cellulose fibres, and parchment paper, made by passing it through a sulphuric acid bath). These technologies consume a lot of energy and have become unsustainable with the steady rise in energy costs in recent decades [35].

Chemicals are an important building block for the diversity and functionality of consumer products and materials [36,37]. They can change the colour and brightness of the paper as well as its strength and water resistance. They are also used to change the material properties and improve the optical and printing properties of the paper. Therefore, the raw materials from which waste paper and board are made are likely to contain heavy metals such as zinc, lead, cadmium and chromium, as well as other chemical additives [38,39].

2.2. Paper Additives

As already mentioned, paper products contain many additives that ensure the special utilisation properties of the paper. In the paper mill, the paper is coated to improve the surface properties. During processing, most of the graphic and packaging papers are subsequently printed and then partially varnished or coated. Throughout its life cycle, paper comes into contact with many different materials, which results in a range of contaminants. The main ingredients used in paper and cardboard packaging materials for their barrier properties are based on plastics, glass and metals [40,41]. Barrier properties of a material can be defined as the protection of food commodities inside the package by preventing the entry and/or exit of different penetrants such as moisture (water vapours), oxygen, carbon dioxide, greases, and oils (*Figure 5*). The use of these ingredients as a barrier in paper-based packaging makes the recycling process challenging and increases the overall cost of recycling [42,43]. The impurities generated during paper production at higher temperatures can become sticky and thus lead to disturbance in the process of recycling. All non-paper components that can form sufficient adhesion and cohesion can be a source of sticky impurities (e.g. resins from wood, coating binders, printing ink binders, coatings, impregnations, adhesives) [44]. Synthetic polymers such as polyethylene (PE), polypropylene (PP), and ethylene vinyl alcohol (EVOH) have been widely used to improve the overall barrier attributes of paper-based packaging [45].

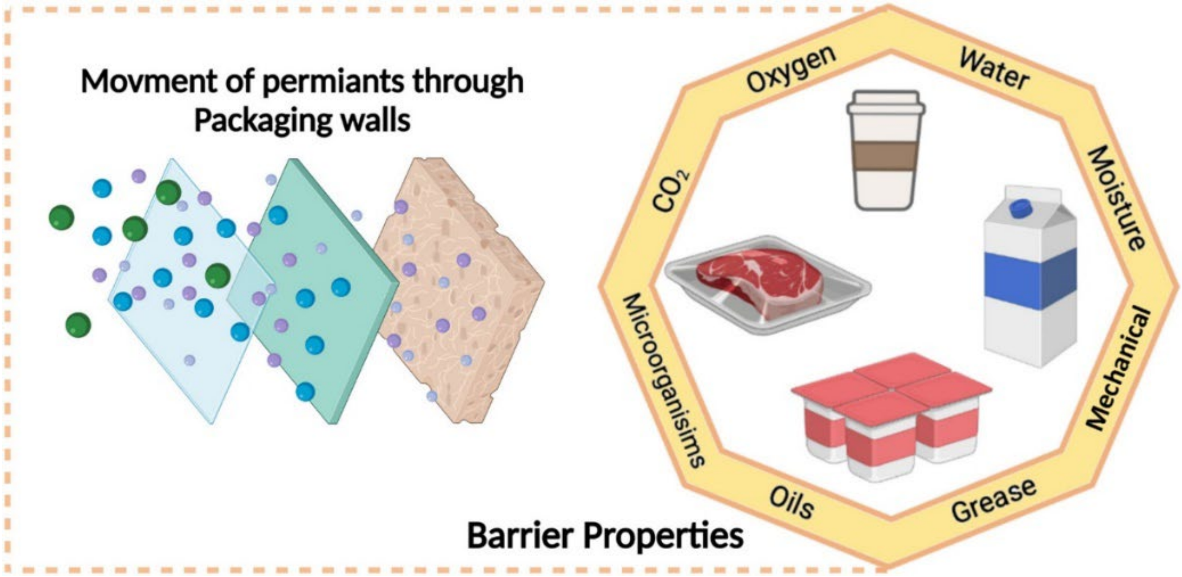

Figure 5. Barrier properties of a material [24]

Additives and fillers ensure the special usage properties of the papers, while coatings and varnishes improve the surface properties of the papers. Additives such as zinc oxide and zinc sulphide are used as fillers and coating materials and cadmium metal has been used to increase the cohesive strength of paper. Calcium carbonate is added as a filler to avoid quality degradation during ageing by adjusting the pH value. Therefore, zinc, lead, chromium, cadmium, etc. can be expected as contaminants in the paper [46]. In addition to minerals and additives, recycled paper can also contain various materials such as staples, laminated covers, plastic wrapping, printing inks, thick adhesive layers, etc. [4]. In general, printing inks are the most important non-fibre components that can be found or contained in recycled paper. In addition to printing inks, lacquers or overprint varnishes can also be found as non-fibre components. They are uncoloured forms of printing inks and can be used to provide additional gloss and protective properties to the print and substrate [47]

2.2.1. Adhesives in Paper Industry

Adhesive bonding is a process that adheres two surfaces permanently joined together by the application of an adhesive material. While this process is well understood for compact materials, it becomes more complicated for porous fibre-based materials such as paper and paper board. The strength of an adhesive bond depends on various physical properties of both the paper and the adhesive. Adhesives come in many forms and types, and the choice depends on the substrates to be bonded, the machinery used in the process and other factors, such as the possible requirement for food-safe materials. Most adhesives are applied using specially designed machinery, making the selection process even more complex and requiring adhesives with specific properties that match the operating parameters of the equipment. In some cases, adhesives are developed specifically for a particular machine type or model [6].

Adhesives play a crucial role in the production of almost all goods, especially mass-produced articles. To form complex finished products, most paper and cardboard products are joined together using adhesives. They are used in a variety of paper bonding applications, from corrugated box construction and lamination of printed sheets to packaging materials for all types of consumer goods. An adhesive film should have a much higher intrinsic strength than paper or cardboard (fibre tear) than the substrates to be bonded, and it must have properties that do not interfere with recycling. In other words, the adhesive should be easy to remove and it is of great importance to choose adhesives that do not interfere with the recycling of primary materials. In recycling processes that take place at high temperatures (e.g. glass or metal recycling), the influence of adhesives, which usually consist of organic polymers, can be neglected. In the field of low-temperature recycling technologies, the question of whether an adhesive is recycling-friendly or not can only be answered by knowing its application and the recycling process. If the recycling processes are known, it is easy to select suitable adhesives [44]. It is important to choose the appropriate adhesives to avoid problems in the recycling process, which will lead to the competitiveness of the recycled material [15,48,49].

Until the 1940s, naturally derived materials such as paste and glue were used as packaging adhesives. Nowadays, starch and casein-based adhesives, natural rubber latex, polyvinyl alcohol emulsion, and petroleum wax in combination with polymers and tacky resin are used as adhesives [17]. Various types of adhesives are used, such as water-based adhesives, acrylic emulsion-based synthetic adhesives, polyvinyl acetate-based adhesives, polyvinyl alcohol-based adhesives and combinations of polyvinyl acetate and acrylic emulsion, depending on the requirements of the packaging material. With increasing global energy problems due to the scarcity of petroleum resources, the industry's focus has shifted to the development of sustainable adhesives that are cost-effective and renewable [6]. Various biomaterials are currently used as sustainable adhesives, including starch, cellulose, soya, itaconic acid and chitosan. In addition to the mentioned bio-based adhesives are made from naturally occurring materials such as animal or agricultural products such as starch, cellulose, protein, casein, animal glue gelatin, natural rubber, etc. Adhesives with 100% solid content such as heat seal adhesives and hot melt adhesives have also been used in the paper industry, which is environmentally friendly [9].

As mentioned before, adhesives are used to bond cardboard surfaces together and create a permanent bond. The bond created by adhesion must be strong enough to be processed as soon as the paperboard leaves the glueing machine. Therefore, the adhesive must be selected taking into account the absorption properties, such as the setting time, which is adapted to the paperboard and the process of the glueing machine. The main criterion for the successful glueing of packaging products is almost always 100% fibre breakage when the adhesion line is separated. The type of adhesive typically used to bond coated paperboard substrates is based on the chemistry of polyvinyl acetate (PVAc) together with water and other functional additives. To achieve uniform fibre tearing when bonding coated/uncoated substrates, the base sheet of both substrates must always be the weakest link when separating. The mechanism for achieving a uniform fibre tear under the coated surface is such that adhesive carriers (mainly water) must penetrate the fibre matrix to cause "fibre bond breaks" (weakening), which in turn trigger a fibre tear. During this process, adhesive particles are trapped in the pores of the substrate where they "harden" and form a permanent bond. [50]

2.2.2. Coating

Coating is the treatment of the paper surface with clay or other pigments and/or adhesives to improve print quality, colour, smoothness, opacity or other surface properties [6]. Due to their porous nature, controlling the wetting and barrier properties of paper and board is extremely important to maintain the shape and mechanical properties of the packaging by limiting fibre swelling and loss of quality of the packaged goods. These functions can be achieved by internal sizing or further treatment of the paper with external barrier coatings. The coating is an aqueous suspension containing several components: Pigments (kaolin clay, calcium carbonate, talc), binders (starch, SB latex, acrylate latex, CMC), additives (lubricants, insolubles, optical brightening agents) and water. The most important colour component of the coating is the pigment, which can be mineral or synthetic. The proportion of pigments in the dry coating is around 80-95% of the weight of the coating. Another important component of the coating colour is the binder, which has the task of adhering the pigment particles to the cardboard surface and, also binding the pigment particles to each other. Normally, the proportion of binder in the dry coating colour is about 5-20% of the weight of the coating [14].

Today, barrier coatings for paper are usually based on fossil or synthetic polymers, which dominate the current market due to their low cost and easy availability. These polymers include polyolefins (polyethylene), waxes, ethylene vinyl alcohol (EVOH) and polyvinylidene chloride (PVDC), which provide a significant barrier to water and oxygen penetration in food packaging. However, they are penalised by limited fossil oil reserves, the poor recyclability of coated papers and the lack of biodegradability, which adds to the environmental and economic concerns [51]. Biobased coatings and adhesives for paper and cardboard are being developed to improve the barrier potential [32,43,52]. Cardboard grades are usually coated with white mineral pigments to achieve good print quality and improve substrate properties. There is a great demand for paper with a very smooth printing surface [6]. The coating fills the cavities and covers the highest sitting fibers on the surface of the cardboard substrate, but defects cannot be completely hidden. The coating reduces the penetration of the ink into the board; consequently, the ink does not spread too much and the printed image remains clear and sharp.

A variety of coatings are used in multilayers to achieve specific functional properties. Many are applied from aqueous dispersions that are dried and coalesced into a thin film. Others can be dissolved in an organic solvent. In the past, toluene was often used, but this has now been replaced by safer, less odorous compounds such as methyl ethyl ketone, ethyl acetate and ethanol. Proprietary mixtures of various chemical components are used extensively to achieve the desired functional properties therefore the chemical descriptions are inevitably somewhat simplified. The list of the most common coatings is listed below.

Protective coatings

They are primarily used to protect surfaces from mechanical damage, but can also give gloss and heat resistance and protect against chemical corrosion. There are three main systems: a) non-curing systems based on nitrocellulose resin (NC), b) thermosetting systems where high temperatures are used in the drying tunnel of the coating machine to induce thermosetting reactions. For example, epoxy resins made from epichlorohydrin and bifunctional phenols can be cured with phenolic, amino or anhydride resins – or combinations thereof. c) Systems that cure at ambient temperatures include those based on reactive polyurethane systems similar to those described above for adhesives.

Heatseal coatings

Polymers and copolymers of vinyl acetate, vinyl chloride, styrene and modified polyolefins such as ethylene acrylic acid (EAA) and ethylene vinyl acetate (EVA) are generally used. Blends are often used to ensure a good bond, both to the substrate, e.g. aluminium foil, and to another material to which it is to be bonded, e.g. a polystyrene-based pot. Blending can also be used to achieve peelable seals. In some applications, polyester-based systems are increasingly being used to provide a chlorine-free option.

Primers

Polyethyleneimines are widely used, but there are also other systems, e.g. EAA dispersions or reactive polyethyleneamines, which are used to prime aluminium foil for PE extrusion coating.

Multifunctional coatings

PVdC coatings based on copolymers of vinylidene chloride with vinyl chloride or alkyl acrylates can be tailored to the desired properties such as gloss, heat sealability and barrier against moisture, gases and odours or flavours. Aqueous dispersions of polymers and copolymers based on acrylic and methacrylic acid are used to produce acrylic coatings that offer similar functionality but a much lower barrier.

Cold seal coatings

Aqueous dispersions containing blends of acrylic resins with natural rubber latex are used for the production of coatings that can be sealed into each other without heat and only with pressure. Synthetic rubber is used for a small proportion of the formulations. Release lacquers Polyamide-based release coatings are applied to the opposite surface to prevent such cold seals from sticking when wound under pressure in the reel [53].

2.2.3. Multilayer Packaging

Today's needs can no longer be met by traditional packaging and more advanced and creative forms of food packaging are required. Modern society demands modern solutions, especially in the area of food safety and therefore also food packaging [54]. The reason for using multilayer materials is the need to design packaging that combines different functional requirements [53]. The structure of multi-layer packaging consists of two or more substrates that are joined together with adhesive usually based on polyvinyl acetate (PVAc) [51]. TCurrently, multilayer packaging represents the largest proportion of non-recyclable packaging and accounts for around 20% of all flexible packaging. The reason for the high proportion of multilayer packaging is that by combining different materials, customised property profiles can be created with low material consumption.

Defined barrier properties against oxygen, water vapour, light or loss of aroma enable a longer shelf life and thus the current form of food trade and reduce food losses. As the different polymers/materials are in many cases immiscible, this packaging cannot normally be recycled. In addition, current recycling systems are aimed at recycling mono-materials, which is why multilayer packaging is sorted out and sent for incineration [55].

Multilayer print designs are also commonly used in commercial food packaging to attract consumers [56]. Most existing multilayer packaging generally consists of three to twelve layers. However, in the case of food packaging, multilayer packaging consists of three to seven layers as shown in *Figure 6*. Apart from the number of layers, multilayer packaging can be categorised into flexible packaging (such as films) and semi-rigid packaging (such as beverage cartons and multilayer bottles) depending on the degree of flexibility [57].

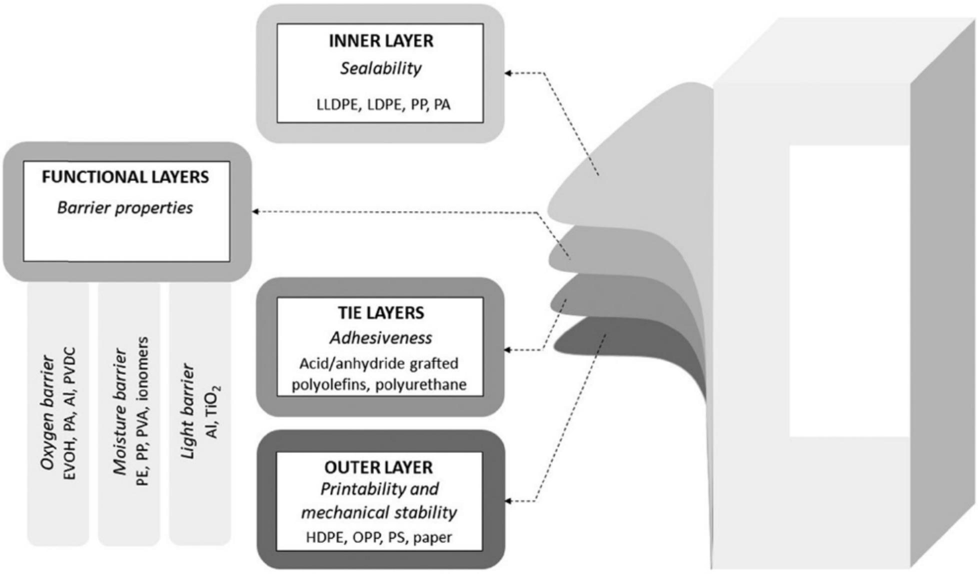

Figure 6. Multilayer packaging structure [57].

In flexible packaging multilayer film composite production, traditional dry lamination causes significant VOC emissions and high energy consumption. The production of flexible packaging composite film, especially for retort pouch food packaging, must fulfil consumer expectations and comply with environmental regulations while guaranteeing product yield and quality. The article by Joachimiak-Lechman presents a comparison of the alternative manufacturing processes of laminates used for packaging in the food industry. The aim of the study was to assess the potential environmental impact of the production of two-layer laminates with a patented polypropylene film Metallyte™ 28UBW-ES and the previously used pap/LDPE/Al/LDPE multilayer laminates. Both the life cycle assessment method and the life cycle cost method were used in the study. The difference in costs between the manufacturing processes analysed is relatively small. In terms of environmental impact, however, there is a clear disparity in favour of the two-layer laminates [58].

3. FOOD AND PHARMACEUTICAL PACKAGING

Packaging is known as a protective outside layer, whose main objective is to protect and preserve food from potential physical, chemical, microbiological, or other risks that may ultimately affect its quality and safety. Food packaging plays an important role in protecting mentioned risks along the food chain. Because of their similar properties food and drugs undergo the same rules and regulations [59]. Packaging is essential to deal with external influences such as odours, impacts, dust, temperature, light, and moisture to which food is exposed. Without packaging, food handling would be costly and inefficient. Carefully selected materials ensure the desired mechanical, physical, and thermal resistance as well as optical, barrier and antibacterial properties. Packaging is a necessity in society today, as virtually all consumer goods on the market are packaged with a limited amount of packaging material [60].

Packaging can delay the deterioration of products, preserve the positive effects of processing, extend shelf life and maintain or increase the quality and safety of food. The aforementioned functionalities captured the attention and interest of scientists and industry, so they got involved in innovating packaging products [61]. Packaging offers protection against three major classes of external influences: chemical, biological and physical. Chemical protection minimises changes in composition caused by environmental influences such as gases (typically oxygen), moisture (increase or decrease) or light (visible, infrared, or ultraviolet). Many different packaging materials can form a chemical barrier. Biological protection provides a barrier to microorganisms (pathogens and spoiling agents), insects, rodents, and other animals, preventing disease and spoilage. In addition, biological barriers maintain conditions that control senescence (ripening and ageing). Such barriers function through a variety of mechanisms, including preventing access to the product, preventing odour transfer, and maintaining the internal environment of the package.Physical protection guards against mechanical damage and absorbs the shocks and vibrations that occur during distribution. Physical barriers are typically made from cardboard and corrugated board and resist impact, abrasion and crushing, so they are often used as shipping containers and as packaging for delicate foods such as eggs and fresh fruit. Appropriate physical packaging also protects consumers from various hazards [16].

Food spoilage is usually caused by oxidation, microbial spoilage, and metabolism, which can be influenced by environmental pollution and other factors such as temperature, humidity, light, physical damage, microorganisms, odours, shocks and dust. Different categories of food have different storage and transport requirements. For example, the preservation of fruits and vegetables requires the reduction of respiration and transpiration rates, which can usually be achieved by controlling humidity, temperature, light, gas environment (O_2, CO_2, ethylene) and so on. For dairy products such as milk, cheese and cream, oxidation and microbial growth must be avoided, and therefore external conditions such as oxygen, light and humidity must be carefully considered. Meat products suffer from the problem of discolouration, which can be prevented by vacuum packaging or modified atmosphere packaging. To ensure the safety and quality of meat products, environmentally friendly biopolymer packaging is often used [62].

Packaging also provides consumers with an initial product identity before they decide to buy. The style and design of the packaging can also improve the image and acceptance of the product, so the design of food packaging plays an important role. Consumer demand is also changing and now includes packaging as diverse as active and smart packaging. These packaging systems interact and react to the food packaging environment by releasing certain substances into the packaging space or intercepting them from the packaging space and extending the shelf life of food. Such innovative packaging is practised in part to increase sales in a highly competitive environment.

The interaction between food and its packaging is a decisive factor, especially when the food comes into contact with the packaging material. This contact results in the penetration of gases and volatile substances, moisture, microorganisms and other low-molecular compounds. This interaction between food and packaging materials is considered an exchange between food, packaging and the environment and can have an impact on food quality, safety and/or the integrity of the packaging. The main objective of food packaging is to protect food from external environmental influences, but the interactions between food and packaging can also affect the quality and/or safety of food.

3.1. Food Contact Material

Many factors should be considered when selecting packaging materials, including cost, product quality and the ability to maintain product freshness. Some common materials used for food packaging are plastics, paper, glass and metals. A variety of plastics are used for rigid or flexible food packaging. Packaging materials today also include laminates, which have been developed by systematically integrating materials with different properties to improve the functionality of the final material. Primary packaging is main package used to hold food being processed; secondary packaging combines primary packages inside one box; tertiary packaging combines multiple secondary packages into one pack [63].

Cellulosic paper and paper-based materials are one of the oldest and most widely used food contact materials (FCMs), often used for dry food packaging [64,65]. An FCS is "any substance intended for use as a component of materials used in the manufacture, packaging, transport, or holding of food when the use is not intended to have a technical effect on that food" [16]. Approximately 47% of total paper and paperboard produced in 2000 was used for packaging purposes [17]. Their use is favoured not only by economic factors (paper packaging is extremely cheap), but also by their safety of use.

The food contact materials witch have recycled paper in their composition, must meet several basic safety criteria. This means that recycled paper that comes into contact with food must not lead to the migration of substances that could endanger human health. The U.S. Food and Drug Administration (FDA) regulates packaging materials under Section 409 of the Federal Food, Drug, and Cosmetic Act. The primary method of regulation is the food contact notification process, which requires manufacturers to notify the FDA 120 days before placing a food contact substance (FCS) on the market for a new use.

The food processing industry selects the packaging material according to the requirements of the food product, taking into account factors such as heat sealability, processability, printability, strength, barrier properties, cost efficiency, sustainability and legal requirements.

White top linerboards used for food packaging are largely made from recycled paper. Either virgin fibre pulp or medium and higher quality waste paper is used to produce the white top plies of multi-ply test liners. In Croatia, the white top ply is usually made from deinked pulp, which comes from wood-free paper printed using the offset process. This means that the layer that is intended to come into direct contact with food is made from deinked pulp. On the other hand, office printouts are mostly used for the production of tissue paper, writing and printing paper, paperboard, and newsprint, but have rarely or never been used as a fibre raw material for the production of the top ply [66].

As mentioned before, paper and paper-based products are porous and offer little resistance to the migration of chemical compounds [67,68]. Therefore, FCMs are made from raw materials and so-called intentionally added substances (IAS), which extend shelf life but also improve production, stability, and mechanical properties [7,8]. IAS include monomers, prepolymers, antioxidants, lubricants, surfactants, and UV stabilisers [36,37]. FCMs may also contain unintentionally added substances (NIAS). The recycled paper contains a total of more NIAS than virgin paper. Several studies have been conducted to compare fresh and recycled paper fibres. The results showed that recycled fibres contain more mineral oils, impurities and overall, more NIAS. Targeted analysis is generally performed for predicted NIAS, while an untargeted or full-scan screening method is used to detect and identify unpredicted NIAS [12,69].
Identification of these substances is often difficult, sometimes impossible [70,71]. For NIAS screening, substances can be divided into three main groups: volatile, semi-volatile and non-volatile [72,73]. Volatile compounds can be detected by gas chromatography (GC) with flame ionisation detection (FID) or mass spectrometry (MS) [73]. Universal detectors are favoured to ensure the detection of the largest possible quantity of substances [74].
NIAS can come from reaction by-products, oligomers, degradation processes, chemical reactions between the packaging materials and the food or as impurities from the raw materials used in their manufacture. NIAS are not only found in plastics but also in non-plastic FCMs (e.g. paper and board, coatings, adhesives, printing inks, silicones, glass, and ceramics). NIAS include all substances that are not added for technical reasons during the production of FCMs. They have different sources and can be categorised as by-products, degradation products and contaminants. NIAS can enter the FCM supply chain at any level, e.g. during the chemical synthesis of raw materials as well as during production, transport, and recycling. Awareness of NIAS as a food safety issue has increased in recent years as sensitivity in chemical analysis has increased and potentially hazardous chemicals migrating from FCMs are being incidentally

identified. As many FCMs have a high chemical complexity, a complete characterisation of all NIAS is currently unrealistic. It is estimated that tens of thousands of substances migrate from FCMs, making the identification of NIAS that may be of concern a challenge. Although more and more NIAS are being identified over time, not all these known substances have been risk assessed. Other NIAS have been identified by chemical analyses, but their structures are still unknown, so that no conclusions can be drawn about their safety. The last group of NIAS are substances that remain completely under the radar because they are not detected by any of the analytical methods used. NIAS can be predicted based on knowledge of chemical processes, manufacturers experience and conditions of use. Such substances can then be relatively easily identified and quantified by targeted chemical analyses. However, many other NIAS cannot currently be predicted. They can either be detected by non-targeted screening methods or remain completely unknown. In *Figure 7*. the various sources of NIAS are described and selected examples are given that illustrate the current state of knowledge, but also the difficulties in analysing these undesirable compounds.

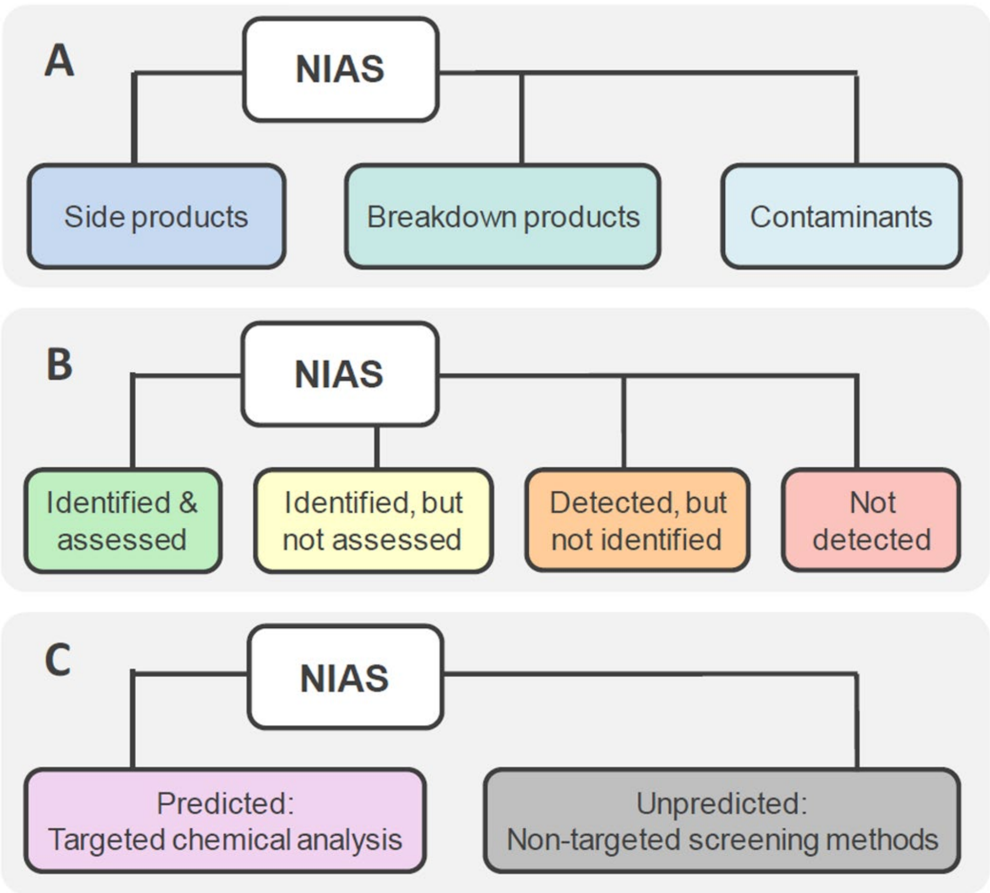

Figure 7. Shows origins and categories of non-intentionally added substances (NIAS) [71]

In paper and board, NIAS are mostly components of printing inks, adhesives, sizing agents and surface coatings. Some migrating substances can be predicted because they were used in the manufacture of the FCM or react in known ways. However, many reaction products and impurities are unpredictable, and the amounts of NIAS present in the FCM and potentially migrating into food are usually even less predictable. The presence of potentially toxic NIAS is often unknown to the manufacturer itself [71]. This makes it difficult to assess their safety when they are exposed [70]. Materials such as paper and cardboard are porous and offer little resistance to the migration of chemical compounds, and direct contact with the food is not a prerequisite for migration [67].

High-resolution precision mass spectrometry (MS) is a valuable tool for the analysis of non-target substances, including screening and chemical identification, and LC-Orbitrap MS is used to screen suspected migrating compounds in paper food packaging [75,76]. Universal detectors that ensure the detection of the largest possible amount of substances are preferred [73,74]. However, there is no single technique for the assessment of trace metals in materials or their migration, and usually several of them must be combined [67]. In packaging that comes into direct contact with food, contaminants can migrate, that is, chemical compounds from the structure of paper and cardboard packaging can move into the food [77]. This requires a comprehensive analysis of all ingredients that can reach toxicological concentrations in food [67].

3.1.1. The Differences between Pharmaceutical and Food Packaging

Food and pharmaceutical packaging are both equally difficult to design. Food packaging is far more diverse than pharmaceutical packaging, while pharmaceutical packaging operates in a much more regulated environment. Some understanding of the differences is useful and even essential for crossover products such as medical foods. As more and more foods are enhanced with ingredients that may cause a change in the body, or as manufacturers make claims about the benefits of foods that may be considered drug-related by the FDA or Federal Trade Commission, regulation and the amount of testing required for product approval increases exponentially. These products are supplied in familiar food containers that are appropriate for the type of food. However, the containers must comply with pharmaceutical regulations that were not required when the product was pure food. It is becoming increasingly difficult to determine whether the packaging must comply with food or pharmaceutical regulations. Food and pharmaceutical packaging follow two different paths in packaging development, which not only have many similarities but also major differences.

The design of the pharmaceutical packaging is very important. Most medicines are packaged in opaque containers that do not allow an easy view of the contents. There is no sensory component such as odour or taste. This makes the labelling of medicines even more important. The contents must be precisely described on the label. Even with slight differences in shape and the use of embossed symbols or imprints on the outside of a tablet, it can be difficult to distinguish between several medicines that are part of a patient's treatment. Colour is helpful, but the most important method of distinguishing one tablet from another is labelling. Accurate labelling is important for patients taking any prescription medication to achieve a therapeutic outcome [78].

3.1.2. Pharmaceutical Packaging

Pharmaceutical packaging (PhP) as a collective term can be defined as the science, art and technology of enclosing or protecting products for distribution, storage, sale, and use, including printed materials used in the finishing of a pharmaceutical product. It serves as an economical means of identification, presentation, protection, and information [79]. PhP is a complex group of products consisting of different materials, including various plastic polymers, glass, paper, steel, cardboard, and metals. The packaging of pharmaceutical products is a wide-ranging, comprehensive, and multi-layered task. In practice, it involves information and knowledge from a variety of scientific disciplines, including chemistry, engineering, materials science, physical testing, sales, marketing, environmental science and regulatory affairs, to name but a few [78]. Depending on the need, there are numerous requirements for PhP materials, but ensuring the quality and function of pharmaceutical products is of paramount importance. Requirements include protection from external influences such as humidity, oxygen, exposure to light, temperature fluctuations, biological contamination, and mechanical damage. Regulatory requirements, such as identification through labelling and product information, are also necessary to ensure safe use, prevent the marketing of counterfeit medicines and ensure that users receive the correct information [80]. In conclusion, a medicine is not a medicine without the right packaging.

The pharmaceutical packaging sector is undoubtedly a very specialised sector that requires attention when promoting the circular economy, as health and safety are the main aspects of decision-making. Legislation, a lack of information or interaction between stakeholders and rigid practices have been identified as barriers to circular product design. The studies have shown that similar barriers and drivers for promoting the circular economy can be found in different but comparable sectors, so we can learn from previous studies focussing on the circular economy in the food industry. The pharmaceutical packaging sector could follow the circular economy decisions made in the food packaging sector on the potential use of chemically recycled materials that come into contact with food. When converting packaging practices to circular processes, the causal relationships along the value chain need to be understood. The environmental impact of circular solutions must also be analysed. A shift in thinking is needed in all value chain activities by increasing interaction between stakeholders and investing in the quality of environmental information. In recent years, the sustainability of the pharmaceutical industry has gained increasing attention from consumers, policymakers and organisations. Health and pharmaceutical products are widely used, but their impact on the environment is still largely unknown [81]. Pharmaceutical companies are increasingly prioritising becoming more sustainable by developing medicines that have the same medicinal value but have less impact on the environment. The choice of appropriate packaging for medicines also influences the emissions added to the environment [82]. Concern for implementing sustainability practises in the development of new delivery systems, new products that pose less environmental risk, waste recycling, reducing water consumption, greener manufacturing methods and recyclable packaging have increased attention to this issue. The sustainability of the pharmaceutical industry has also attracted the interest of scientists from various disciplines such as chemistry, engineering, and environmental sciences. Even business administration and management are looking at this topic. The management of pharmaceutical companies is becoming increasingly complex. The need to ensure environmental, economic, and social sustainability and to control costs has led to the introduction of an economic logic into the management of sustainability issues. Analysis shows that environmental sustainability has moved to the centre of management studies, particularly concerning cleaner production, a green supply chain, green materials, and sustainable human resources management [83].

It is well known how the development of the pharmaceutical industry has impacted people's lives because it contributes to the overall well-being in terms of health and quality of life of individuals and because it is one of the most important economic sectors at a global level [84].

The pharmaceutical industry is responsible for the research, development, manufacture and commercialisation of medicines, vaccines, and treatments for common and rare diseases. Therefore, the pharmaceutical industry has attracted increasing attention due to its impact on the sustainability of its activities [85]. The environmental pollution generated by medicines throughout the life cycle of the product is just one of the issues that require reflection on how the pharmaceutical industry is changing its behaviour in the conduct of its business [83]. In general, the amount of packaging has increased in the wake of increasing political pressure to reduce waste and increase material recycling. The targets and solutions are expected to be found in the circular economy; however, there is a lack of literature on recyclable pharmaceutical packaging [86].

Pharmaceutical packaging material is a collection of various components that surround the pharmaceutical product from manufacture to use. The use of packaging materials has increased significantly over the years. This has led to a growing volume of packaging waste and waste disposal challenges. Like many other types of packaging, PhP is typically disposable. All investments and materials used for packaging are lost if they cannot be recycled. Packaging is one of the most important product value chains for which urgent action is needed to change unsustainable practices. The EU has a strong focus on packaging, which is defined as one of the sectors where the potential for circularity is high, and the targets and measures for recycling and recyclability of packaging are ambitious. These measures include a reduction in both packaging itself and packaging waste, increased use of reusable and recyclable designs and a reduction in the use of complex packaging materials. Studies have shown that well-designed packaging can lead to significant environmental benefits by reducing losses along the value chain. Furthermore, the environmental benefits resulting from the recycling of packaging often outweigh the benefits of energy recovery. However, the environmental benefits depend to a large extent on various factors, starting with the material in question. PhPs can be complex materials due to their many requirements and the required recycling technology is still at an early stage. Therefore, it is necessary to identify the key activities in the value chain to gain insight into the possibilities of increasing the recyclability of PhPs. In addition, the value chain is subject to constant scrutiny by parties within the value chain, such as companies, or by parties outside the value chain, such as the EU [86].

The issue of environmental safety is a major concern for the packaging industries of both industrialised and developing countries. Pharmaceutical packaging companies are among the leading innovators in the industry, as evidenced by recent technological advances. The current trends are the result of many challenges that the industry is constantly facing. Packaging is a science that is constantly evolving and makes an important contribution to the success of the pharmaceutical industry [87].

When studying pharmaceutical packaging, the standard division into three categories (primary packaging, secondary packaging and tertiary packaging) has its own special requirements.The primary packaging is the first packaging envelope that comes into contact with the pharmaceutical form or device. It must be designed in such a way that it does not interact with the medicinal product and provides adequate protection for the medicinal product. This type of packaging includes blister packs and strip packs. Secondary packaging is consecutive wrapping or packaging in which medicinal product packaging such as cartons and boxes are stored to group them. Tertiary packaging is used to handle and transport pharmaceuticals from one location to another, such as containers, drums, etc. [87].

There are five main functions of pharmaceutical packaging.

1. Containment - The containment of the product is the most important function of pharmaceutical packaging. When designing high-quality packaging, the needs of both the product and the manufacturing and distribution system must be taken into account. This requires that the packaging does not leak or allow diffusion or permeation of the product, that it is strong enough to hold the contents during normal handling, and that it is not altered in its final dosage form by the ingredients of the formulation.

2. Protection - The packaging must protect the product against all adverse external influences that could affect its quality or efficacy, such as light, moisture, oxygen, biological contamination, mechanical damage and counterfeiting/adulteration.

3. Presentation and information - The packaging is also an important source of information about medicinal products. This information is provided to patients through labelling and package inserts.

4. Identification - The printed packaging or its printed additional components serve both identification and information purposes.

5. Convenience - Convenience is related to the use or administration of the product, e.g. single-dose eye drops, which both eliminate the need for preservatives and reduce the risks of cross-infection by administering a single dose [88].

3.1.3. Pharmaceutical Packaging Design

Pharmaceuticals can be separated into two segments: medications with a prescription (Rx medications), and medications without a prescription, or so-called „over-the-counter medications". This subdivision of products also determines the design of the packaging. Of all the packaging used today, the printed folding boxes occupy the largest segment. Printed pharmaceutical cardboard packaging, with all its specific details, represents the greatest challenge for folding cardboard manufacturers alongside Tobacco folding boxes. The reason for this is the very high protection standards and the speed of the filling machine's products also dictates the design of boxes.

For a long time, the packaging of Rx products was purely a matter of necessity and was not given importance in terms of marketing. The reason for this is that the doctor who writes the prescription seldom sees the packaging of that medicine himself, and the patient uses what is prescribed. Today, pharmaceutical companies pay more attention to the appearance of their products, which they also present to users as part of the product. Thus, the packaging can not only protect the product from external factors but also make it easier to consume, protect against misuse, or offer the final, important piece of information.

Above all, pharmaceutical packaging must convey a sense of unparalleled precision, cleanliness, and safety. It should not resemble any other form of packaging making it difficult to replace it with another product. A worldwide regulation was implemented, stating that non-pharmaceutical items must not be packaged in a blister-like form (like chewing gum used), to prevent youngsters from unintentionally opening the packaging and potentially endangering their lives. Since conventional advertising methods might not be used to promote this kind of medication, a visually appealing and recognizable graphic design serves as an indirect means to promote the company itself. The pharmaceutical industry recognizes the needs of individuals with disabilities, Braille is now printed on a wide variety of packages, making it much easier for blind people to live independently.

The marketing of prescription medications is highly specialized and distinct from the marketing of any other type of product, particularly when we take into account that these items cannot be promoted through the use of the mainstream media. Given that customers do not make the final purchasing decision, it is crucial to stress that the manufacturer targets a very specific professional group of people, namely doctors and pharmacists, rather than a broad spectrum of consumers. The advent of over-the-counter (OTC) pharmaceuticals is, therefore, a novel and creative marketing strategy that has allowed the pharmaceutical industry to finally gain access to the mainstream media and, consequently, much more direct access to end consumers without the need for physician participation.

Packaging is a powerful marketing tool for promoting OTC medications, and while their quality and safety are governed by the same laws and regulations, there are some variations in their design and sales strategy. An effective OTC design must effectively market the product, make it appealing to the user, educate them about its advantages, and provide instructions on how to use it correctly and warnings if it is not used properly.

Designers are allowed to use all the advantages of modern graphic technology. Solutions with five to seven colours are available, as well as foil printing and a variety of box designs that were previously only found in the cosmetics industry. The role of design in this group of pharmaceuticals is basically the same as that of design in foodstuffs. A well-designed product has to draw potential customers by standing out from the multitude of similarly priced competitors [89].

3.1.4. Construction of Pharmaceutical Packaging

Pharmaceutical packaging belongs to the group of advertising-commercial boxes that are made of cardboard with one or more layers of different fibre compositions, either recycled or pure cellulose. The usage of specific carton types, which cannot be altered without prior consent and machine testing, is obligatory for box manufacturers. Foldable boxes are particularly important for pharmaceutical packaging, including small boxes (for medicines, cigarettes, toothpaste, etc.) and large boxes (for detergents or liquor bottles, etc.). The material used for the production of foldable boxes is mainly cardboard. It can be uncoated, coated or coated on one side (uncoated on the side to be printed, while the other side is finished with a layer to achieve the properties required for packaging certain types of products). According to some authors, foldable boxes include simple boxes, protective/transport boxes, decorative boxes and commercial boxes. They are printed in printing houses, primarily using offset printing, followed by gravure printing, digital printing, letterpress printing and, to a lesser extent, flexographic printing [90]. For the production of food, confectionery, cosmetics, tobacco and pharmaceutical packaging the grammage range of 160-450 g/m^2 is used [14]. They have the largest share among the paper-based packaging categories and are usually quite easy to recycle in standard paper mills [91].

There are three basic box types commonly used in the manufacturing of pharmaceutical packaging shown in *Figure 8*. as follows:

1. standard boxes (adhesive is applied to one spot)
2. boxes with semi-automatic bottoms (adhesive is applied to one spot)
3. auto bottom boxes (adhesive is applied to three spots) [89]

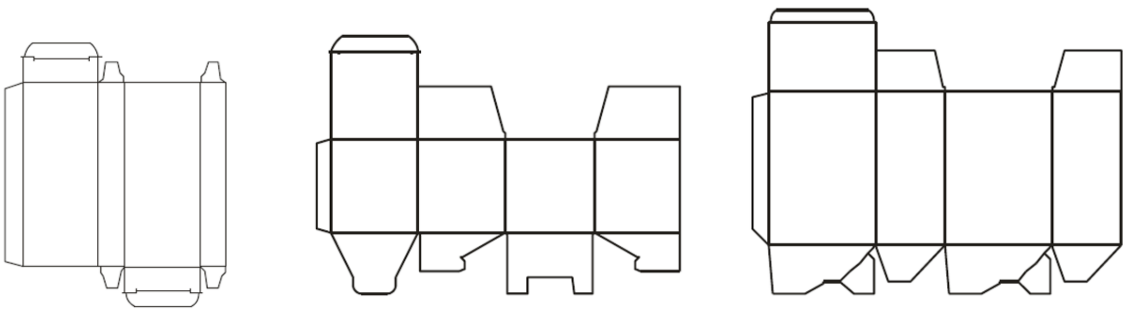

Figure 8. Three basic box types commonly used in the manufacturing of pharmaceutical packaging [89]

3.1.4.1. Printing Machine

A machine for printing paper packaging for pharmaceuticals typically involves several key components and processes. Before printing begins, the design for the packaging is prepared using graphic design software. The first step is to make the box structure according to the specifications and design, which serves as the stamping tool. This includes the layout of text, graphics, and any required information such as dosage instructions, ingredients, and branding elements. This software contains most of the internationally accepted box shapes, providing the technical drawing. Basic dimensions are then entered into the program, together with weight and thickness of the cardboard that will be used to construct the box. The design is then transferred onto printing plates. These plates are typically made of metal or polymer and contain the negative image of the design. The plates are mounted onto the plate cylinders of the printing press. Registration marks are used to ensure precise alignment of each color plate. Each color in the design requires a separate plate. The ink fountain is prepared by loading the required ink colors and adjusting ink keys to control ink flow. The ink is transferred from the fountain to the ink rollers. It is also crucial to adjust the dampening system to control the amount of moisture applied to the plate for maintaining proper ink-water balance and achieving optimal print quality.

The press is configured according to the specific requirements of the job, including the type of paper, ink colors, and dimensions of the packaging. The paper stock, usually in the form of large rolls, is fed into the printing press. As the paper moves through the press, each printing cylinder applies a different color of ink to the paper according to the design. Ink is transferred from the ink rollers to the printing plates, which then transfer the image onto the blanket cylinders. The image is then offset from the blanket cylinders onto the paper. Modern printing presses often use techniques like flexography or offset printing for high-quality results. After each color is applied, the printed paper passes through a drying system to ensure that the ink sets properly. This may involve heat drying or UV curing depending on the type of ink used. Registration adjustments may be made as needed to correct any misalignment.

Once all colors are printed and dried, the paper may undergo additional finishing processes such as coating, varnishing, or laminating to enhance its durability, appearance, and protective properties. After finishing, the printed paper is cut into individual pieces and creased along the necessary fold lines to facilitate folding into the final packaging shape. This may be done inline as part of the printing press or on a separate machine. The creased paper is folded and glued to form the final packaging structure. This may involve complex folding patterns depending on the design of the packaging. It can undergo additional finishing processes such as coating, varnishing, cutting, folding, and binding, depending on the specific requirements.

Throughout the printing process, quality control measures are implemented to ensure that the printed packaging meets the required specifications in terms of color accuracy, registration, print clarity, and adherence to regulatory requirements for pharmaceutical packaging. Once the packaging is produced, it undergoes thorough inspection to detect any defects or imperfections.

This may involve visual inspection as well as automated systems for detecting errors such as misprints or incomplete folds. When inspected and approved, the printed packaging is stacked, bundled, and prepared for delivery to the pharmaceutical companies for use in packaging their products. Overall, the process of printing paper packaging for pharmaceuticals is highly sophisticated and involves precise coordination of various technologies and processes to ensure the production of high-quality, compliant packaging materials [89]

3.1.4.2 Offset Printing

Offset printing, also known as offset lithography, is a widely used printing technique with which the majority of recovered paper is printed. It's called "offset" because the ink is not directly transferred onto the paper, but rather from the plate to a rubber blanket, and then onto the paper. It is known for its high-quality results, especially for large print runs, as it can maintain consistent image quality and colour accuracy, and it supports various paper sizes and types, making it versatile for different printing needs. Offset printing is a method of mass printing in which the images are transferred onto metal plates on rubber blankets or rollers and then onto the print medium [92,93] The print medium, usually paper, does not come into direct contact with the metal plates. This extends the service life of the printing plates. In addition, the flexible rubber easily adapts to the surface of the print medium, meaning that the process can also be used on media with a rough surface such as canvas, fabric or wood. The main advantage of offset printing is its high and consistent image quality at a competitive price. In 2013, offset printing covered around 60% of the total printing demand in Croatia [94]. Conventional offset printing inks are highly deinkable under alkaline conditions [22]. Offset printing is a chemical-intensive process that generates many types of waste [95]. The most important environmental aspects of offset printing are use of non-renewable resources (mineral pigments in paper, mineral oils in inks and solvents, metal plates, metal and plastic in equipment), use of toxic or harmful substances (additives in inks and adhesives, biocides in fountain solution), VOC emissions (from mixing and drying of inks, evaporation from fountain solution, evaporation from cleaning solutions, blanket washes), toxic waste (ink waste, waste from cleaning solvent waste), regenerative waste (paper, unacceptable prints), energy consumption (production facilities, ink drying, transport) and transport emissions (supply chain of paper, inks, varnishes, delivery of printed matter, transport of waste) [5].

3.1.4.3 Printing Inks

Printing inks are colourful, complex mixtures whose main purpose is to convey a message, provide protection and give a decorative effect to the substrate to which they are applied. They are usually composed of colorants (pigments or dyes), binders (resins, oils or solvents), solvents (oil or water-based) and additives (chelating agents, antioxidants, surfactants, biocides, etc.). Pigments are dispersed solids that are insoluble in the support material and are used almost exclusively in printing processes. They are usually finely ground powders. Compared to pigments, dyes are soluble and have lower lightfastness and water resistance [22]. One of the main functions of printing inks is to adhere to the printing substrate and remain there throughout the life of a print. The adhesion of inks to paper plays an important role in many industrial applications and can be characterised indirectly by thermodynamic analysis of the paper substrate or directly by paper laminate or adhesion tape peel testing [96].

Printing inks are formulated for the individual printing processes, both for classic (offset, flexographic, screen, gravure) and digital printing (electrophotography, inkjet, laser). They must have certain rheological properties so that they can be transferred to the printing plate and then from the printing plate to the substrate (e.g. paper). After the ink has been applied to the substrate, the binder dries and binds the colorant to the substrate under the running conditions of the printing press. The drying of printing inks must take place as quickly as possible and can be achieved by physical (evaporation) and chemical (oxidation, radiation-induced curing) means or a combination of both [22]. The printing process is associated with environmental problems due to the consumption of energy and material resources. The impact on the environment depend on the printing technique and the materials used. Offset printing techniques generate the following waste: Substrates, inks, fountain solution, metal plates, volatile organic compounds, biocides, varnish and energy [97]. Printing inks can also significantly affect the quality of newly produced materials, such as recycled paper. If the printing inks are not removed from the pulp, recycled paper not only has poor optical and mechanical properties, but can also become problematic due to the presence of toxic substances. It is therefore very important to carry out a toxicological assessment of new materials. Some types of prints show significant problems during recycling. Most problems occur when using water-based flexographic inks, inkjet inks, UV-curable inks and liquid toners. Ultraviolet (UV) inks are often used for food packaging printing due to their ease of use, space saving and fast drying [56]. The life cycle assessment found that the sheetfed offset printing process is the largest contributor to the environmental impact of most printed products (print 52%, paper 31%, ink 17%), with the dominant impact being acute ecotoxicity [98]. While the economic standard of living has improved, environmental pollution has increased accordingly, which has led to a rapid increase in emissions of volatile organic compounds (s), which are one of the main causes of air pollution. Air pollution affects not only the human respiratory system, but all body systems, including the cardiovascular and reproductive systems. Chemicals can in many cases be replaced by other agents that have less impact on the environment and health. UV inks are designed for suitable drying with LED UV lighting and therefore reduce electricity consumption [5]. Unfortunately, there have been a number of health warnings related to food contamination from photoinitiators in UV-curable inks [99]. Photoinitiators are used in UV inks to initiate polymerisation of binders and accelerate curing. The results of several studies show that photoinitiators migrate significantly from recycled cardboard into dry foods such as cookies, cereals, pasta, rice, noodles or oatmeal [100,101] as well as into fatty foods such as milk powder, infant formula, and juice drinks [102,103]. The ink compounds migrate from the outer surface (printed outside of the packaging) to the inner surface [104], which can lead to kidney failure, endocrine disruption and lung cancer [105]. In order to manufacture usable products from recycled paper, the ink printed on the paper must be removed. The deinking processes used by the paper industry have worked very efficiently in the past, but this is beginning to change with the increasing use of inkjet printing on an industrial scale. Research by Bolanča et al. has shown that the removal of ink using conventional offset printing methods is very effective, as these inks are hydrophobic and can agglomerate to a size of 50–200 microns, leading to strong interactions between the ink, surfactants, and air bubbles [5]. The problem occurs if the printing inks are not completely removed during paper recycling. For example, printing inks can contain heavy metals (copper, lead, zinc, chromium and cadmium) and solvents. In addition, paper recycling produces a sludge that contains short fibres, fillers, printing inks from the deinking process, extractive substances and deinking additives which should be properly disposed of [106,107].

4. HEAVY METALS

Heavy metals are among the most important pollutants and can be found everywhere in the environment [108]. They accumulate in plants from the environment such as soil and air and are transferred to animals via the food chain [109]. Humans ingest heavy metals through the consumption of food contaminated with heavy metals, which can have an impact on human health. Of particular concern is exposure to hazardous chemical product components that are found in a variety of everyday consumer goods [110,111]. The health relevance of substances depends on the toxicity of the substance and the extent of exposure. At a very low levels of exposure, adverse health effects are generally considered negligible [112–114]. However, some chronic diseases have been linked to exposure to certain chemicals that migrate from food packaging. A number of studies have determined heavy metals in different food categories. The European Food Safety Authority (EFSA) has published scientific opinions on the contamination of Pb, Cd, Al and As in food (Lee et al., 2019), and in 2001, the U.S. Environmental Protection Agency (EPA) compiled a list of twenty hazardous substances, with Arsenic (As), Lead (Pb), Mercury (Hg) and Cadmium (Cd) at the top of the list [115]. Pb is considered a potential carcinogen and causes a number of serious health problems, caused both by short and long-term exposure to it. Pb is also considered a carcinogen and has been linked to lung, kidney, bladder and skin cancers. High blood pressure and cardiovascular disease are closely linked to arsenic exposure. Cd is one of the most dangerous heavy metals and was first described as the cause of Itai disease in Japan in the 1950s. Long-term exposure to high concentrations of Cd leads to skeletal damage and has been classified by the International Agency for Research on Cancer (IARC) as carcinogenic to humans. Aluminium (Al) is considered a strong neurotoxicant and can cause irritation. Several studies have shown that there is a significant association between Al in drinking water and Alzheimer’s disease.

In recent years, there have been several research efforts to provide recommendations for the safety of chemicals used in specific packaging and to provide a science-based basis for the development of risk management strategies [75,102,116–119]. Apart from the general requirements, there is still no harmonised legislation in the EU for paper-based packaging materials intended for direct contact with food or for the use of recycled paper fibres in contact with food. In the absence of a specific directive, the paper and board food packaging supply chain relies on the national paper and board legislation published in some European countries. The existing regulations define the chemicals that are permitted in the production of paper and board and set limits for various contaminants in the end products. The Packaging and Packaging Waste Directive (94/62/EC) provides the framework for limiting the presence of heavy metals in packaging [120]. This directive limits the concentrations of lead, cadmium, mercury and hexavalent chromium in packaging materials to a maximum value of 100 ppm (mg/kg) for the total content of all four metals combined. This standard applies to all packaging within the EU and imported packaging material. The recommendation of the German Federal Institute for Risk

Assessment (BfR) for paper and board for food is considered the most reputable regulation in Europe for paper packaging [121]. In addition to the applicable national laws and existing regulations for paper and board for food contact, there are also guidance documents such as the CEPI guidelines for food contact of paper and board materials and articles developed by the European paper and board manufacturing industry as well as the converting industries and other associations. These documents are aimed at manufacturers of paper and board materials and are intended as a guide to compliance with Regulation (EC) No 1935/2004. However, the guidance documents are not legally binding. In the United States, packaging regulations are often state-specific, such as California's Proposition 65, which requires labeling of products containing chemicals known to cause cancer, birth defects, or other reproductive harm [122]. Although Proposition 65 does not specifically target packaging, its provisions affect packaging materials used for products sold in California. The circular economy is the primary goal of socially responsible and sustainable businesses, but the aforementioned standards emphasize the importance of knowing the composition of recycled raw materials and their suitability for health.

4.1. Contaminants in Food Packaging Materials

The use of recycled paper is by far the highest in the packaging industry. In the production of food packaging, recycled paper is often preferred to paper and board made from virgin fibres.
However, as a significant proportion of recycled fibres have been printed at some point, the suitability of using recycled fibres from printed sources in food contact applications is still being assessed in terms of health safety [121]. In other words, the recycling of printed papers can introduce contaminants into the food packaging material, which under certain conditions can transfer to the packaged food, posing a health concern for consumers.
Studies on paper-based food packaging materials published in the last two decades have identified a variety of substances that may be present in recycled pulp [123–126].

In paper recycling, these substances remain in the solid matrix and thus end up in new products based on recycled fibres. Binderup et al. were among the first authors to provide a long list of potentially harmful substances in recycled paper, including phthalates, solvents, azocolorants, diisopropylnaphthalenes, primary aromatic amines, polycyclic aromatic hydrocarbons and benzophenone [123]. To maintain high paper recycling rates with a satisfactory quality of recycled pulp, it is essential that paper recycling processes are highly effective in removing inks and toxic components. In addition, it is of great importance to look for ways to reduce the occurrence of harmful contaminants and minimise their migration into the packaged food [127].

4.2. Heavy Metals in Food Packaging

Heavy metals are considered as one of the most important parameters in the evaluation of paper packaging. Chemical contamination can occur in all phases of the material and product life cycle. Numerous toxic chemicals such as printing inks, phthalates, surfactants, bleaching agents and hydrocarbons are introduced into the paper during the development process [17,128], therefore food packaging made of paper must be tested for various contaminants including aromatic amines, benzophenone, polyaromatic hydrocarbons, plasticisers and heavy metals [46]. For example, to improve the structural and surface properties of the paper pigments are added. Zinc sulfate is used to increase the opacity of special papers, while zinc oxide is sometimes used to make photocopy paper [129]. Zinc and cadmium pigments are additives that give paper fluorescent properties and increase the cohesive strength of paper surfaces. Zinc is also commonly used in papermaking for fine art applications when white pigments are used [130]. Metals such as copper and aluminum are used for engraving on various packaging [131]. Additionally, the quantity of toxic metals can increase when treating corrugated board packaging using dyes that dissolve in water and an acidic platform without previous surface treatment [129]. The main sources of heavy metals are dyes, which mainly consist of conventional inks and pig-ments, as well as spot inks and Pantone Matching System (PMS) inks [77]. Inks are considered to increase the content of Cd, Pb, Zn and Cu [132]. In general, confectionery is packaged in brightly coloured packaging to attract customers as it is likely to be consumed by young children. It was concluded that most pigments used in printing inks are based on metal compounds of Zn, and Cu, including Pb and Cr, which is why they are already banned for food packaging in some countries [133]. Green and yellow packaging can contain compounds such as lead chromate, lead sulfate and lead oxide [134].

4.3. Migration

The paper and board food packaging chain is currently facing the challenge of combining the desire for food safety with the desire for a circular economy. As mentioned afore, various chemical substances are found in foods during different phases of the supply chain; these include micronutrients, flavorings, antimicrobials, antioxidants, pesticides, and mycotoxins.

Also, additives such as plasticizers, monomers, and oligomers found in the packaging materials could transfer to the foods upon contact during processing or packaging; this transfer of chemical compounds between the food and packaging is termed "migration". This interactive phenomenon could result in alterations in the quality and also the safety of the food, and flavor may change owing to sorption of aroma and the transfer of undesirable components from the packaging material to the food. Understanding the migration mechanism is crucial for estimating food deterioration when using synthetic polymer-based packaging [63].

When recycled paper is used in food packaging, it must fulfil many specific requirements of European legislation. This means that recycled paper must not release any substances that could pose a health risk to consumers. Over the years, studies have been published on the migration of these toxic chemicals from paper, cardboard and plastic into food and food simulants [135–139]. Overall, the results of the studies show that migration from paper and cardboard depends on the structure of the packaging samples, the chemical nature of the migrants and the time and temperature conditions of the migration tests. Therefore, the use of recycled fibres can be problematic as long as the scientific basis of migration from cellulose fibres is still poorly understood, as are the properties that allow chemicals to withstand washing processes during recycling and subsequently migrate into packaged dry food. However, the economic and environmental benefits of paper recycling are considerable, and recycled paper is used in direct contact with dry foods such as flour, cereals, sugar, salt, rice and pasta [117]. Studies on recycled materials have identified a variety of heavy metals as well as other chemical additives that may be present in recycled pulp [38], most of which originate from printing and converting processes carried out when the paper was previously used. Muncke et al. reported that more than 10,000 chemicals are intentionally used in the production of food contact materials [140]. These substances remain in the solid matrix during paper recycling and thus end up in new products based on recycled fibres [141]. In addition, certain types of paper and board types in household waste paper contain relatively high concentrations of chemicals such as mineral oil hydrocarbons, phthalates, phenols, polychlorinated biphenyls and toxic metals (Cd, Co, Cr, Cu, Ni and Pb) (Götze et al., 2016). Biedermann investigated potential migrates from recycled cardboard used to produce boxes for packaging dry foods and showed that most of the components detected were not natural components of the fibres, but residues of printing inks, adhesives, impurities and other chemicals in the recycled material [142]. Another study showed that higher concentrations of metals in paper and cardboard waste are not necessarily due to cross-contamination from other waste materials, but may originate from certain paper and cardboard components such as printing inks and fillers [143].

Mohammadpour et al. detected harmful metals such as Pb in high concentrations in most pastry packaging made from recycled paper [144]. In another study, different types of packaging and levels of heavy metals such as Al, As, Ba, Cr, Co, Ni, Pb and V were analyzed, which in some samples exceeded the permitted concentration [145]. The work of Sturaro et al. aims to investigate the migration of chemicals from recycled paper and cardboard packaging into dry food and to provide guidelines for the packaging industry [139]. Chang et al. developed a new Liquid Chromatography Mass Spectrometry (LC-MS/MS) analytical method. This method was found to be effective in the rapid analysis of photoinitiators with a high degree of reliability [56]. The ICP-MS method developed for determining the mass fractions of chemical elements (Al, Ba, Fe, Mg, Mn, Pb, Sr, Zn) in paper samples proved to be linear and precise. Values of relative measurement uncertainty ranged from 7.7% to 13.6%, and the used approach allows for improving the quality of data and decision-making [146]. The work of Canellas et al. using GC-MS and UPLC-QTOF for migration study has shown that the identification of compounds in the adhesive is a crucial step, as NIAS found in the adhesive migrated into the food simulant. Therefore identification of NIAS is crucial for materials that come into contact with food. It was shown that the choice of materials that make up the multilayers can be decisive, as the migration values depend strongly on the material that the compounds have to pass through [147].

4.4. Assessing Metal Traces in Paper

There is no single technique for assessing metal traces in materials or their migration, and usually several must be combined [67]. It must be emphasised that migration is usually performed with different food simulants: liquid (e.g. 95% ethanol, 3% acetic acid) and/or solid (Tenax®). The analytical methods used to analyse food simulants depend on their chemical properties. Naturally, if a target analysis is performed, other analytical methods such as gas chromatography coupled with a flame ionisation detector (GC-FID), high performance liquid chromatography coupled with an ultraviolet detector (HPLC-UV) and high-performance liquid chromatography coupled with a fluorescence detector (HPLC-Fluorescence) as well as ultra-high performance liquid chromatography coupled with triple quadrupole mass spectrometry (UPLC-QqQ-MS) can also be used [54] High-resolution, accurate mass spectrometry (MS) is a valuable tool for analysing non-target substances, including screening and identification of chemicals in food packaging materials. LC - Orbitrap MS is used for suspicious screening of migrating compounds in paper-based food packaging [75,76]. The content of chemical compounds in printed packaging is usually measured using gas chromatography (GC), a sophisticated technique for analysing substances [14].

5. RECYCLING

The market for recycled paper has existed worldwide for thirty years (Bandara & Indunil, 2022). More than 40% of the total paper production is based on the use of secondary fibres [148]. Despite the environmentally friendly component associated with the use of paper, there are still environmental concerns associated with paper production and its recyclability. The production of pulp, paper, paperboard and wood products contributes significantly to climate change emissions, chemical pollution and energy consumption [149], however manufacturers and recyclers of paper packaging are committed to using the best environmental practices [150–152]. The recycling of waste paper requires 28–60% less energy than paper production from virgin fibres, as most of the energy is consumed in the pulping of the virgin fibre raw materials. The main goal of conventional paper recycling is to decrease the impact of paper use on forests by disassembling and reassembling paper [153]. The paper industry generates tonnes of waste of different compositions and types at various stages of production, disposal and recycling. Landfilling and open dumping of waste is not a favourable choice, so incineration is the best disposal option for the paper production and recycling industry due to the associated energy recovery. Alternative disposal options such as pyrolysis, composting, gasification and reuse as building material are good, but still need to be optimised. The additives used in paper production also affect the recycling of waste paper, but recycling is necessary as it reduces the pressure on virgin timber for the supply of pulp [17]. Recycling conserves natural resources, saves landfill space and reduces air pollution from incineration. The energy savings in the production of recycled paper amount to 70% compared to the production of virgin fibre paper. In addition, paper recycling saves large amounts of water. It also leads to a reduction in gas emissions and water-polluting emissions [154]. Due to the positive impact of paper recycling on the environment, the amount of paper recovered for reuse has increased significantly over the last two decades. In 2022, up to 70.5% of all paper and board consumed in Europe was recycled, making it one of the most collected and recycled materials in Europe. According to

CEPI Key Statistics, almost 60% of the paper collected was used for recycling in the production of packaging paper and board [152].

The use of paper for recycling as an additional resource has increased worldwide and represents an alternative to traditional paper recycling. Furthermore, it could potentially address the problems associated with traditional paper recycling [155]. However, paper can be recycled up to seven times because the fibres lose their quality after a certain number of recycling cycles and become unusable[156] and the quality of recycled paper is sometimes inferior to paper made from virgin pulp due to the short length and lower tensile strength of the fibres [157]. During the recycling process, the fibres are subjected to mechanical processes each time, which leads to a deterioration of their mechanical properties. While recycling plays an important role in the circular economy, it is also necessary to extend the products lifespan. The inadequacies of the current waste management system highlight the importance of reducing the demand for paper packaging material (PPM) by implementing more aggressive waste prevention programmes and strategies [158].

It is generally recognised that using recycled rather than virgin material is a more sustainable solution, as recycling contributes to sustainability and the circular economy [159]. For this reason, a large proportion of recycled fibres are used in food applications as primary or secondary packaging in many European countries. The use of recovered paper in white grades has experienced significant growth since the 1970s with the introduction and spread of flotation deinking [4]. Recycled paper can be made from a mixture of post-consumer paper, reject paper from paper mills, pre-consumer paper such as trim paper and surplus paper from printers, which may contain virgin fibres. The recycled office paper grades are mainly used for the production of tissue paper, writing and printing paper, paperboard and newsprint. It is worth noting that recovery rates for office waste paper are very low (13.1%), although they account for more than 30% of the total recovered paper collected [160]. Blasius & Manoiu investigated the suitability of using sorted office paper grades in the production of white top linerboard to minimise production costs. According to their study, sorted office paper could serve as an alternative substitute pulp and is one of the most valuable papers in the paper industry as it contains a high proportion of virgin fibres. White top linerboards used for food packaging, are largely made from waste paper. Virgin fiber pulps or recovered papers of medium and higher grades are used to produce the white top plies of multiply test liners [161].

Currently, 90% of pulp is made from wood. Paper production requires about 35% of deforested trees, which is equivalent to 1.2% of the Earth's total economic output. Recycling one tonne of newsprint saves one tonne of wood, while recycling printing or copy paper saves more than two tonnes of wood [46]. The depletion of non-renewable resources has led to an increasing trend towards more efficient utilisation of waste newspapers (WNPs) as a raw material for various applications. As typical waste biomass materials, WNPs account for about 7% of municipal solid waste and contain low-cost cellulosic material [21].

The paper mills that produce recycled paper or cardboard have numerous cleaning systems, including sorting machines and, for graphic papers, deinking systems. After a series of cleaning processes, the paper suspension is subjected to a separation of impurities based on density, size or shape [162]. Refining is necessary when using virgin fibres to produce cardboard or paper. This is a process in which various chemical agents, colours, varnishes and coatings are used to achieve a high-quality printed surface. A print substrate produced in this way potentially contains fewer pollutants than a print substrate made from recycled fibres. As discussed before, recycled fibres may contain certain levels of chemical contaminants, including phthalates, alkanes, ketones and aldehydes [163]. According to CEPI (Confederation of European Paper Industries), the defined targets for the recycling of paper-based packaging are 75% by 2025 and 85% by 2030. CEPI has proposed a list of recommendations for improving the recyclability of paper packaging in the paper recycling process [164].

5.1. Deinking

In recent years, the use of recycled fibres has significantly increased the need for a more fundamental understanding of flotation deinking. Indeed, most of the equipment in today's plants needs to be renewed and a more fundamental understanding of the process should lead to a simplification of operations so that higher fibre concentrations can be treated [165]. Due to the increasing tendency to print on certain products such as cardboard, the deinkability of packaging is of great importance. Recycled paper, which is usually printed with various types of imprints, does not always have positive deinkability tests. In the deinking process, the printing ink and other contaminants must be removed from the recycled mixtures. Deinking by flotation is particularly efficient with conventional offset and gravure inks when hydrophobic inks are present [166], and recent studies have shown that recycling repetition can have a positive effect on deinkability efficiency [167]. According to Faul 81% of offset prints, mainly newspapers and magazines, achieved a positive assessment of their deinkability [4]. Deinking is of great importance in the production of graphic papers due to the high demands placed on the optical properties of the end products. It is also becoming increasingly interesting for packaging grades [168]. Paper and board are easily reusable after their production, but their different applications with non-paper components make the process difficult. Coatings, varnishes, laminates, adhesives, etc. can affect their deinkability or have an impact on the environment [169]. Research must address the environmental issues arising from the uncontrolled consumption and management of non-biodegradable materials [170].

The use of recovered paper in the production of graphic papers depends on the deinkability of the printed paper, which makes deinking crucial and one of the most important key factors to achieve the desired properties of recycled pulp. Packaging grades and mixed grades are usually recycled without deinking. For graphic paper grades, deinking is common practise, with the exception of some high quality grades with little or no printing ink [168].

Studies have shown that deinking depends on many factors, such as the quality of the paper collected for recycling, the type of printing process, the properties of the inks, the age of the printed product and the climatic conditions during its life cycle [166,171,172]. The surface energy of the paper and the wetting properties of the ink can also significantly affect the deinkability properties of the inks due to the adhesion of the ink to the paper [173]. It is successful for most of today's printing inks and printing technologies found in recovered paper [174].

Various deinking processes can be used for the production of recycled fibres, depending on the quality of the paper to be recycled and the desired quality and requirements of the pulp produced. The most common deinking process is flotation to remove printing ink from waste paper. In this process, chemicals and air are used to form bubbles that can capture and remove the ink particles in the pulp suspension [168]. This makes the method expensive and leads to environmental pollution due to the release of contaminants, so choosing the right deinking chemicals is often a trade-off between cost and performance [175].

5.1.1. Flotation Process

Successful flotation consists of a few steps. The detachment of the ink particles from the hydrophilic cellulose fibres in an alkaline solution, followed by the adhesion of the hydrophobic

ink particles to the surfaces of the air bubbles and the removal of the flotation foam with the ink particles from the flotation cell [176,177]. This process can be improved by high shear conditions, chemical effects and moderate temperatures in the range of 55 to 70 ˚C. This leads to swelling of the fibres, which is promoted at a high pH value (8–11). The chemicals required in the flotation steps, such as collectors, frothers, pH regulators, dispersants, etc., are usually added in the pulping stage to ensure good mixing and dissolution [165,178].
The detached ink is removed from the pulp by washing, flotation, screening or centrifugal separation (i.e. deinking step), followed by bleaching to improve the visual appearance [179]. This step usually contributes the most to chemical pollution in wastewater streams [180,181]. The conventional deinking process uses a large amount of chemicals such as sodium hydroxide, sodium carbonate, diethylene triamine pentacetic acid, sodium silicate, hydrogen peroxide and surfactants, which lead to hazardous effluent disposal problems. Reduction in chemical consumption and the generation of toxic compounds is required for environmentally friendly paper production [182].

The conventional recycling approach is still a major contributor to energy consumption, climate change emissions and the release of toxic chemicals into the environment. Currently, flotation deinking, where the process works with a fibre consistency of 0.8%, consumes large amounts of water. In order to efficiently remove ink particles by flotation deinking, two conditions must be met: the detachment of the ink particles from the fibre and the removal of the ink by agglomeration of these particles and their adhesion to bubbles. These processes are governed by physico-chemical phenomena and special chemicals are essential for an optimised flotation process. Efficiency gains and environmental benefits can be achieved by replacing and modifying conventional chemical systems and processes. Approaches to increase efficiency and reduce the environmental impact of the above life cycle include the modification of the deinking pulping process [165,183].

5.1.2. Stickies

During the process of recycling paper, a large amount of stickies accumulates and get deposited on the surface of forming fabrics, press rolls, felts and dryers. These compounds come from the raw materials used in paper production and their appearance is enhanced by closed water circuits in the paper mills. The handling of recycled fibres is more complex than when processing primary fibres, as recovered paper consists of a mixture of different fibre types or types of paper. Pollutants and harmful substances that can contribute to the formation of sticky impurities must be removed from the pulp, which will contribute to the quality of the raw material. The term "purity" is usually used in relation to optical, chemical, colloidal, microbiological and processing aspects. Impurities and contaminants are gradually removed based on various separation criteria such as particle size, shape and deformability, density and surface properties of the particles. The sequence of steps depends mainly on the properties of the raw material and the desired properties of the end product. There are two main ways of dealing with stickies when processing waste paper: removing and/or eliminating stickies in the process of sorting, cleaning, flotation and bleaching and preventing the formation of deposits by dispersion, fixation and removal of stickiness [184].

Stickies come from inks and special adhesives used mainly in recycled fibres such as corrugated containers, mixed office waste and old newsprint. They can be present in the pulp, process water and/or final product [148]. Stickies are characterized as a variety of sticky forms that are created during the paper recycling process. The term "stickie" does not imply a specific chemical composition, but is derived from the complex physical and chemical nature of a mixture of organic substances that are tacky, hydrophobic and pliable, have different shapes and are denser than water. They consist of substances such as fatty acids, resin acids, sterols and sterolipids, etc. They also include latex, ink and hot melt adhesives. Paper chemicals can also produce stickies. For example, the addition of talc in the Whitewater system is an effective method of removing stickies by adsorbing the stickies to the mineral particles, but can reduce paper strength due to the addition of minerals [185]. Sticky substances can deform and aggregate with temperature changes, have a wide range of melting points and act as adhesives [148].

Knowing the characteristics of stickies is helpful to solve this problem. The main problems associated with stickies are deposits inside the paper machine and on the paper itself. The first type of build-up occurs in the wires, felts and rolls and leads to lengthy, unproductive cleaning times. Deposits also lead to paper braking, where spots adhere to the moving rollers. Deposits in the paper reduce its quality and lead to stacking problems between sheets. Stickies can be minimised using chemical and mechanical methods [148]. The following mechanical methods are mainly used: Screening, flotation with dissolved air and washing with water. These can be used in combination with chemical methods to optimise the cleaning process. Mechanical methods only remove the larger stickies. Chemical methods are the most important method for controlling micro and colloidal stickies. This control is based on the use of dispersants, adsorbents and various additives whose chemical properties are detrimental for their specific effect on the identified stickies. Most methods passivate the surfaces of stickies by pH and temperature adjustment and make the surfaces clean by preventing the deposition of stickies. The problems caused by the occurrence of stickies are not easy to manage and there is no single effective method to control them. Stickies control is judged by paper quality and the reduction of cleaning procedures. Another important issue is therefore the quantification of stickies and the standardisation of their measurement. There are various methods for quantifying stickies, which can be categorised into methods for measuring quantity, composition or deposition tendency [186–189].

5.1.3. Stickies Size

Stickies can be classified based on their size and their tendency to agglomerate. Therefore, they can simply be divided into microstickies and macrostickies according to their physical size. Microstickies can be further categorised into suspended (20 - 100 µm), dispersed (1-25 µm), colloidal (0.01 - 5 µm) and dissolved stickies (< 0.01 µm). In general, microstickies do not pose a problem for the papermaking process unless they accumulate excessively. However, they have a potential to promote the formation of secondary stickies [190]. Macrostickies are larger and cannot pass through a 100-micron-slotted screen plate. They are usually caused by flocculation of the tacky polymer additives with fibre fines, mineral fillers and so on (Licursi et al., 2016), and are associated with the fibrous fraction of the pulp. Microstickies range in size from over 1 micron to under 100 microns and belong to the fine fraction of the pulp [148].

The build-up of macrostickies in the paper machine system can cause the problems associated with web breaks and long unproductive cleaning times, as the weak spots adhere locally to the moving rolls and build up on wires, felts, rolls and doctor blades. It can also slow down water drainage during the wet-end process and/or block the slots during screening. All this affects runnability in the pulp and papermaking process. It is therefore particularly important to control the formation of macrostickies in paper production from recycled fibres [190].
In the pulping process, the larger macrostickies and other impurity particles of appropriate size can be effectively removed by various cleaning processes. Pulp washing is an effective way for removing small particles with sizes < 10 µm, while flotation is more effective for removing medium-sized particles, 10 - 100 µm. The screening and centrifugal cleaners are used for removing the large ink particles (>100 µm) [191].

The main aim of waste paper recycling is to remove printing ink and other contaminants while retaining the optical and strength properties of the fibres. The quality of the recycled fibres depends on the success of the recycling process and decreases the higher the collection rate of recovered paper and the more “marginal” paper fractions are collected [107,143]. Poor fibre quality of recycled paper can lead to problematic situations in various end uses of paper, e.g. printing, writing and packaging. Therefore, it is very important to evaluate the paper quality parameters of recycled paper to ensure the satisfaction of various end users. Above all, the permanence and durability of recycled paper should be measured to understand the paper quality [154].

The International Association of the Deinking Industry (INGEDE), has developed methods for entry inspection and operates a database for its members. Deinkability depends mainly on the properties of the printing inks and therefore on the printing process. Flotation deinking, the predominant method developed to remove letterpress and rotogravure inks, works well with mineral-based offset inks and dry toners. Flexographic and inkjet ink particles are too hydrophilic and too small for efficient flotation. Cured systems and some toners, especially liquid toners, form agglomerates that are too large for flotation.The recognised evaluation scheme for deinkability, i.e. the removability of inks, uses INGEDE Method 11 for testing. The results are converted into “deinkability scores”. The second product-related quality aspect is the ability to remove adhesive applications. This depends not only on the chemical characteristics of the adhesive, but also on the type of application. The INGEDE database on the recycling behaviour of adhesive applications is much smaller than the one on deinkability. The tests focussed on glued book spines and labels. Glued spines often show sufficient recyclability if they are produced with hot melt adhesives. Of these, polyurethane glues are generally the best option. Labels are much more critical; one of the reasons for this is the low film thickness. Not enough is yet known about how the chemical nature can compensate for this disadvantage. To find out more about how adhesive applications can improve their recyclability, INGEDE has started a survey with some co-sponsors on the recycling behaviour of about 200 printed products with adhesive applications [192].

The effectiveness of the deinking flotation process and the characteristics of the obtained cellulose fibers are determined by measuring the optical properties and determining the distribution of the ink particles using image analysis. The characteristics of the paper samples before and after flotation are measured [193].

CIE whiteness

CIE whiteness refers to the measure of how closely a color matches a perfect white reference under specific lighting conditions. The International Commission on Illumination (CIE) defines standards for color measurement and whiteness evaluation. CIE whiteness is often expressed as a numerical value or index. Paper whiteness is determined according to the ISO 11475:1999 standard - Determination of CIE whiteness, D65/10°, while the degree of whiteness (%) is determined by measuring the reflection of light from the surface of the paper in the visible spectrum. The samples are illuminated using a D65 light source, which represents daylight. Whiteness measurement is particularly important when achieving a specific level of brightness or whiteness is desirable for product quality and consistency [193].

ISO Brightness

ISO Brightness refers to a standardized method of measuring the brightness of paper or paperboard products. It is defined by the International Organization for Standardization (ISO) in ISO 2470-2:2008. ISO Brightness is typically expressed as a numerical value, measured on a scale ranging from 1 to 100.

The ISO Brightness measurement is based on the reflectance of blue light (wavelength 457 nanometers) from the surface of the paper sample under standardized conditions. The higher the ISO Brightness value, the brighter the paper appears. ISO Brightness is an important parameter for various applications, including printing, publishing, and packaging, where the visual appearance of the paper is crucial [193].

Opacity

Opacity in paper refers to its ability to block the transmission of light. In simpler terms, it measures how much light can pass through the paper. A paper with high opacity will prevent more light from passing through, making it less transparent or see-through, while a paper with low opacity will allow more light to pass through, making it more transparent.

Opacity is an important characteristic of paper, particularly in printing applications where it affects the readability and appearance of printed materials. Papers with higher opacity are preferred for packaging materials because they prevent text and images from showing through from one side to the other, improving legibility and visual appeal.

Opacity is typically measured using standardized methods, such as ISO 2471 or TAPPI T425, which involve measuring the amount of light that passes through the paper sample under controlled conditions. Opacity values are expressed as percentages, with higher percentages indicating greater opacity [193].

Fluorescence

Fluorescence in paper refers to the emission of light by the paper when it is exposed to ultraviolet (UV) light. Some paper materials contain fluorescent brightening agents (also known as optical brighteners or optical brightening agents) that absorb UV light and re-emit it as visible light in the blue range of the spectrum. This phenomenon makes the paper appear brighter and whiter under daylight or artificial lighting conditions. Fluorescence in paper is often desirable in applications where brightness and whiteness are important factors, such as printing, publishing, and packaging. Optical brighteners are commonly added to paper during the manufacturing process to enhance its visual appearance [193].

ERIC

ERIC measures the components found in recycled pulp, namely residual color particles that absorb light at 950 nm. These particles are of invisible size but have a significant impact on the appearance of paper. ISO 22754:2018 standard is applied to all types of recycled pulp and sheets made from recycled pulp [193].

The CIE Lab* color space

The CIE Lab* color space is a three-dimensional model used to represent colors, with coordinates associated with the perceptual characteristics of color, corresponding to the theory of opponent colors: light-dark, red-green, and yellow-blue.

The symbol L* ranges from 0 to 100, where 0 represents black and 100 represents white. The symbol a* represents the red-green coordinate, and the symbol b* represents the yellow-blue coordinate. Both coordinates can take positive and negative values.

To put it concisely;

L* represents the lightness ranging from black to white.

a* represents the redness-greenness axis.

b* represents the yellowness-blueness axis [193].

Image analysis

Image analysis can be characterised as any form of signal processing where the input is an image. The result of image processing can be either an image or a set of features or parameters related to the image. In most image processing techniques, the image is treated as a two-dimensional signal and standard signal processing techniques are applied to it. There are several image analysis methods for analysing pulp properties such as macrostickies, different fibre morphology properties and dirt particles [194].

5.2. QUALITY OF DEINKING PULP AND PAPER

There are few studies on the deinking of pulp and pulp quality as a food contact material. Hanzer et al. investigated whether deinked office paper grades can be used as an alternative fibre source for the production of white topliner board for food packaging. The hand sheets formed after each recycling trial were tested for their suitability for direct food contact. Deinkability was evaluated by calculating the flotation yield, the brightness and whiteness increase of the pulp, the ink elimination factor, the determination of the residual ink area and the elimination of the ash content. Food safety was assessed by determining the content of heavy metals (Cd, Pb, Hg and Cr), primary aromatic amines, diisopropylnaphthalenes (DIPN), phthalates and polychlorinated biphenyls (PCB) in aqueous or organic solvent extracts of recycled paper pulp.

Regarding the food safety assessment of deinked pulp, all tested deinked handsheets were found to be suitable for direct food contact [121]. In the work of Runte et al. a method for evaluating the recyclability of packaging products on a laboratory scale was developed. Coarse rejects, flake content, macrosticky area below 2,000 μm and optical homogeneity were tested for packaging products of different categories to evaluate the process efficiency and quality of the pulp after recycling. The test shows a very high variation of results between and within the different product categories, which is due to the product design, which is mainly influenced by the purpose of the packaging product. Folding boxboards showed very good recycling behaviour. Only one of the 37 products tested had a coarse scrap rate above 10%, which is the maximum value for folding boxboard. This product was a folding boxboard for frozen food with a PET film. The average coarse scrap was very low at 1%, and more than 50% of the products in this category had no coarse scrap. The average flake content was low at 3.6% and the product with the highest flake content was only 16%. In the folding boxboard products, the macrostickies could originate from both the recycled paper and the adhesive used. Overall, it was shown that corrugated boxes and folding boxboard are quite easy to recycle in standard paper mills [169]. Canellas et al. used the standard deinking method INGEDE 11 to investigate the suitability of an acrylic adhesive for food packaging. The efficiency of the adhesive is compared depending on the material used, laminated and non-laminated paper samples to analyse the recyclability of existing printed products and possible improvements [195].

As mentioned before, there are two main aspects on which the quality of the recovered paper depends. One relates to the quality of the delivered paper in terms of contamination, moisture and composition. The other is related to the recyclability of the delivered paper products [4]. The first aspect concerns the collection system, handling and storage of recovered paper, which depends on environmental awareness, sorting activities, the price of recovered paper and the impact of printing and converting techniques [196]. It is important for the paper industry that paper and board are collected separately from other recyclable materials, and source-separated waste collection systems are an essential part of increasing resource efficiency. The fractions of waste that are introduced into the paper production process should have as high a proportion of cellulose fibres as possible [197]. In Europe, an additional 18 million tonnes of waste could be collected annually if proven collection strategies were used, resulting in a 13% reduction in greenhouse gas production associated with packaging and packaging waste [151]. The differences in packaging waste collection rates across Europe are large even between municipalities with similar characteristics, suggesting that there is great potential to increase the amount of these materials that can be recycled. There are several methods of collecting paper and packaging waste (PPW), however, the use of these the choice of methods can affect the efficiency of recycling and the quality of materials in the further process [71]. As part of the COLLECTORS project, data was collected on the separate collection of packaging and packaging waste (PPW) by waste collection systems (WCS) in Europe. PPW is collected separately from residual waste where different collection methods are used. There is separate collection, where certain PPW materials are separated from each other before collection, and various co-mingling strategies, where different PPW materials are collected together [198]. Increased collection of PPW must be accompanied by higher efficiency in post-collection recycling. This is achieved through systemic improvements in waste management, including better packaging design and technological advances in the sorting and recycling stages, e.g. by replacing mechanical recycling with chemical recycling [199]. Therefore, Doshi et al. suggest improving communication and collaboration throughout the paper production line, including suppliers, manufacturers and chemists [200]. The sorting of paper for recycling is mainly manual (rarely semi-automatic and automatic) and is diverse. Sorting activities play an important role when low-grade sources are used to achieve higher collection rates. According to Rahman et al. the paper industry can choose a specific sensor-based paper sorting system, such as a stiffness sensor for old corrugated cardboard or containers (OCC), a lignin sensor for old newsprint (ONP), a gloss sensor for glossy paper, a colour sensor for writing paper (WP), etc. [201].

Studies have shown the importance of selective collection of paper and board and the application of advanced sorting technologies and their impact on the quality of recovered paper delivered to paper mills. In addition, they have shown that the use of low-quality recovered paper grades contributes to an increased environmental impact through higher energy consumption and a larger volume of waste generated, resulting in higher emissions to air and water (Vukoje et al., 2020). The use of co-mingling methods can affect the recycling efficiency and the quality of the material downstream due to higher levels of contamination [197]. In addition to collection rates, better closing of material loops depends on the quality of waste materials after collection and sorting.

Most of the collected paper is usually recycled, but around 40% it is contaminated with food or wet. This type of paper is undesirable in paper recycling plants due to cleaning difficulties, which leads to contamination problems [203]. In this form, it is not suitable for conventional recycling methods, but it is suitable for organic recycling [204]. The deinking process is designed to remove printing inks, but not to whiten unbleached fibres, therefore the deinking industry is in favour of separate collection of graphic paper from households to reduce contamination with non-deinkable paper and board.

Although high collection performance is crucial for efficient resource utilisation, improving waste collection systems at source alone is not sufficient to achieve recycling targets. By assessing the environmental impact of current waste management, it can be shown where in the system most improvements need to be made for each material to extend its useful life [205]. The transition to a circular economy has become a priority in the European Union's (EU) vision for sustainable economic growth and global competitiveness [151].

6. WASTE MANAGEMENT

Municipal solid waste (MSW) is the most complex stream of solid waste, consisting of food residues, paper and board, plastics and other components. According to global statistics, 2.01 billion tonnes of MSW are generated annually, with an average per capita waste generation of 0.74 kilogrammes. Waste generation is expected to increase by 19 % in high-income countries and by 40 % or more in low- and middle-income countries. Over-packaging continues to lead to excessive use of materials and energy and thus influences the impact of production and transport processes [42,43]It is estimated that the amount of waste will increase to 3.40 billion tonnes by 2050. Therefore, solid waste management has become an alarming problem in both industrialised and developing countries. Various countries in the world are facing many social, environmental and economic problems due to solid waste generation. The main components of MSW are food, plastics, metals, paper and board, rubber, leather, textiles and hazardous substances. The proportion of paper and board in municipal solid waste is around 20 %. Food is the only product class that is typically consumed three times a day by everyone. Consequently, food packaging accounts for almost two thirds of all packaging waste by volume. Paper-based products, together with kitchen waste, are one of the most common materials in the municipal solid waste fraction and waste with the highest recycling rates. In addition, food packaging accounts for around 50% (by weight) of total packaging sales [16]. The recovery of paper for recycling from municipal solid waste is useful as it can be used to manufacture various valuable products. Recycling paper and cardboard reduces municipal solid waste and thus improves sustainability [44]. Recovered paper is today the most important raw material for the production of new paper-based products. The Waste Framework Directive sets out the necessary measures for EU countries to comply with in order to achieve the following targets. By 2020, the preparation for reuse and recycling of waste materials (such as paper, metal, plastic and glass) from households should be increased to a minimum total of 50% by weight. By 2020, the preparation for reuse, recycling and other material recovery of non-hazardous construction and demolition waste, including backfilling with waste as a substitute for other materials, is to be increased to at least 70% by weight. And by 2025, the preparation for reuse and recycling of municipal waste should be increased to at least 55%, 60% and 65% by weight by 2025, 2030 and 2035 respectively [22].

6.1. Circular Economy

The high recycling rate can be achieved through an effective waste paper collection system. Sustainable waste management is based on maximising the recovery of resources and minimising the final disposal of waste. Material returned to the production process reduces material intake from nature and thus supports the sustainability of natural resources. Resource utilisation must be maintained at a rate that does not lead to long-term depletion. The Sustainable Waste Management Summit points the pathway to economic model called the “circular economy”, as opposed to the linear “take-make-dispose” economic model we use today. Our current lifestyle is to take natural resources to make products and throw it away after use. In a circular economy, material flows in a cyclical pattern, which limits unnecessary waste, and limits intake of resources [206]. Despite the valid arguments, the circular economy has yet to gain acceptance because it is not easy to abandon the linear model that we have been practising. The transition to a circular economy requires a new way of thinking that promotes integrated resource management through policy integration. Recent dialogues have shown that the sustainable management of waste can play an important role in alleviating some of the scarcity problems we face with water, food and energy.

The circular economy is based on the economic advantages created by reducing the impact on the environment and reducing resources. It primarily appears in the literature through threemain actions, the 3Rs rule. The WFD introduced the fourth R “recover” as a 4Rs framework as the current EU waste hierarchy. Scholars extended the R-based circularity framework beyond the 4Rs up to 9Rs [207]. The 9R Framework' includes Recover Energy, Recycle, Reuse, Remanufacture, Re-store, Repair, Reuse, Reduce, Rethink and Discard. The premise of discarding in a circular economy should be kept to a minimum, but one must not forget to mention the existence of that phase. To reduce the negative impact of the rejection phase, it is possible to combine this phase with the energy recovery phase [208]. The mentioned phase does not contribute primarily to the circular economy (due to the impossibility of uninterrupted energy consumption without major losses), so waste could become a raw material for obtaining energy.

From a life cycle perspective, both the waste hierarchy and circular economy consider the whole life cycle of a product, including the pre-use phase, use phase, and postphase [209]. Both have evolved over time to emphasize the design and use of a product before it turns into waste. It is shown in *Figure 9*. Circular economy and waste hierarchy share a joint philosophy, aiming to manage waste by rethinking, redesigning, and repurposing in order to improve the resource effectiveness of a product and to reduce the generation and adverse impact of waste. The minor difference is that the waste hierarchy still allows disposal, while the framework of a circular economy does not.

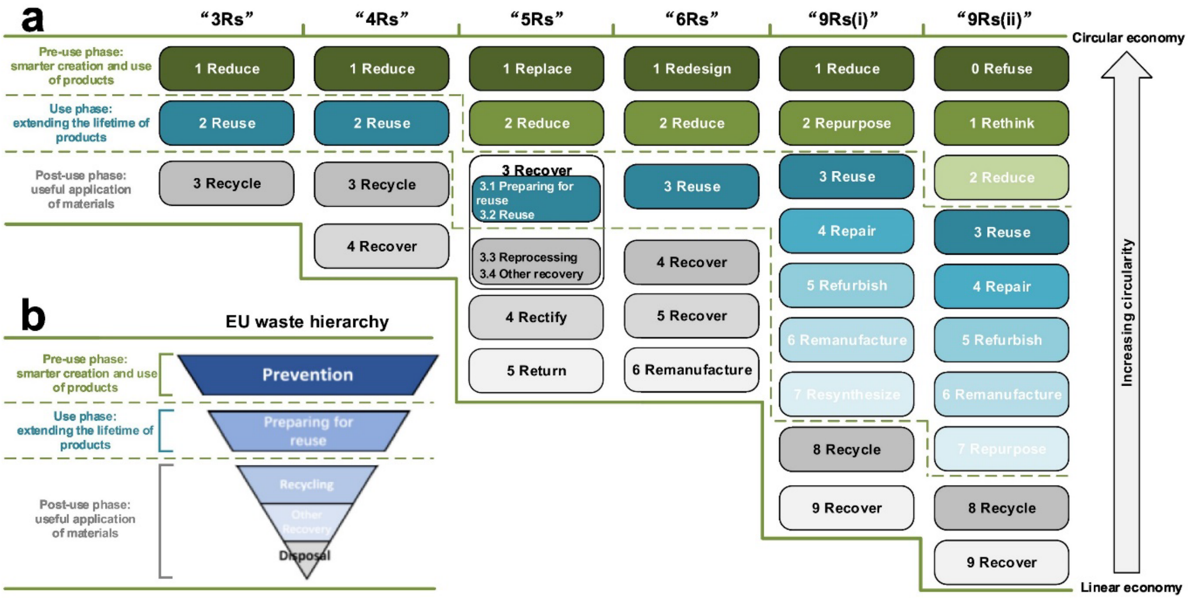

Figure 9. Comparison of circular economy and waste hierarchy philosophy [209].

Other methodologies such as "Circular Economy Product Strategy and Business Model Framework" suggests design and business model strategies to be implemented together [210]. ReSOLVE framework (regenerate, share, optimize, loop, virtualize, exchange), as a basis for some of the best frameworks as backcasting and eco-design for a circular econ-omy (BECE) [211]. Retrospective planning methodology is useful for complex problems in which current trends are incorporated. Such a planning method can increase the probability of solving ecologically complex issues and predict changes with a strong economic impact [212]. Through a holistic approach, BECE (backcasting and eco-design) introduces the postulates of the circular economy into corporate decision-making based on conclusions reached by combining operational and systemic premises [213,214]. The ReSOLVE framework emphasizes technology as the driver of key transformations that will provide new incentives for the adoption of new technologies.

Achieving many of the Sustainable Development Goals (SDGs) set by the United Nations (UN) depends heavily on sustainable waste management. Sustainable development was first described in the Agenda21 document in 1992 and its appendices, which state that the most important challenges for global policy are *improving people's lives and conserving our natural resources in a world that is growing in population, with ever-increasing demands for food, water, shelter, sanitation, energy, health services and economic security*. Moreover, the European Parliament and the Council of the European Union ordered in Directive 2008/98/EC that by 2020, the weight amount of recycled or re-used construction waste must be increased to a minimum of 70%. These challenges can be addressed by renewable resources – reusable, recyclable, available and affordable materials, among other things. One of these renewable resources is paper [23]. The ongoing SDG campaigns can also be used to raise awareness of sustainable waste management and accelerate our transition to a circular economy [215]. Political pressure to reduce waste and improve the material cycle has increased [3]. The idea of a circular economy has been developed as a means to avoid and minimise harmful human activities [86,207,216–219]. Solutions include eco-design, waste prevention programmes and extending the lifespan of products. "Reduce, reuse and recycle" are three important principles [217].

The most important is to prevent and to reduce the amount of all kinds of waste [220]. Most paper-based products have a lifespan of a few days (e.g. newspapers) or a few weeks (e.g. packaging). Reducing the amount of all types of waste streams produced has been mandated by European legislation, either through prevention, reuse or recycling. To improve the recovery of waste, it should be collected separately when technically, environmentally and economically feasible and should not be mixed with other waste or materials with different characteristics. Various options for the disposal of paper for recycling have been investigated worldwide as different problems have arisen. For example, poor deinkability of prints, contamination issues from food, high moisture content of collected paper, etc. Despite the high percentage of paper used for the production of recycled paper and board, different authors make different suggestions, from incineration to bioethanol production and biodegradation in aerobic and anaerobic environments. Research into the environmental and economic impact of packaging sustainability has been stimulated by regulations and market pressures, resulting in numerous guidelines, theories, strategies and tools on packaging sustainability. These have been made available to various stakeholders, including designers, engineers, technologists, marketers and environmental managers in the production, transport and distribution sectors of packaging production [221]. Efficient packaging waste management systems are crucial in light of recent revisions to European packaging waste management legislation, which set ambitious targets. The European regulations aim to tackle the increasing amounts of packaging waste causing environmental problems. It is therefore necessary to develop functioning systems for the management of packaging waste in order to achieve these targets effectively and efficiently. However, the heterogeneity of the various packaging management systems makes it difficult for policy makers, scientists and economic operators to gain a comparative overview. The number of non-harmonised laws in force in different countries, autonomous recycling targets and constant updates are prominent problems that make it difficult to obtain comparable information for research, business and policy [222].

6.2. Sustainability

Since the 1980s, environmental issues have gained dramatically in importance at local and global levels. World Bank scientists in 1992. first used the term environmentally sustainable development, and soon after, the term was used globally. Afterwords in 1995. Goodland coined the term environmental sustainability, which refers to safeguarding human well-being by protecting natural resources. Since then, the term environmental sustainability has gradually gained acceptance and is used by scientists, practitioners, and policy-makers. Environmental sustainability refers to the impact of processes, products and services on the environment, biodiversity, and human health. It is a state of balance, connectivity and resilience that enables human needs to be met without jeopardising the supporting ecosystems and biodiversity [83].

The sustainability approach encompasses energy and material flows, closed-loop systems, cleaner technologies and waste reduction, and economic and social factors. One of the key factors is the development of sustainable products that are defined by their life cycles and that ultimately have a competitive advantage on the market. There are various strategies to reduce all types of waste streams produced, but in general current waste management strategies are shifting from waste disposal to recycling, reuse and recovery [168]. The growing demand for sustainable business practises is also important in the graphic arts industry.

Development and production in a more sustainable way has received special attention in recent years. Packaging products range from single materials with simple designs to complex products containing different materials (cardboard, wooden boards, paper, plastic, etc.). Knowing the environmental impact of packaging products used in a specific production area is not only important for improving the environmental performance of products and/or processes, but also for meeting the demands of the market for ecological/green products [223].

6.3. Life Cycle Assessment

Sustainability and environmental issues are one of the most important topics for strategic corporate and product development decisions. Sustainability in development includes energy and material flows, clean technology, closed-loop systems, quality as well as economic and social aspects. Life Cycle Assessment (LCA) examines the environmental aspects and potential impacts of a product throughout its life cycle, from raw material extraction to production, use and disposal. It is a well-established and widely accepted analytical tool for determining the environmental profile of a product, covering the entire environmental impact of a product, process or human activity, from the procurement of raw materials through production and use to waste disposal. LCA it is the most objective and widely used tool for determining the environmental profile of products with comprehensive approach whick makes it a unique tool in the field of environmental management. It is often used to reduce material and energy consumption and pollution during product design and manufacture. Since the 1970s numerous organisations and standardisation companies have attempted to systematise this type of assessment and make the necessary improvements [224].

Nowadays the ideas of the life cycle approach and eco-efficiency have become increasingly practical and are applied in various sectors of the economy, including the packaging industry [203]. Eco-efficiency analyses in the packaging industry cover both the production of packaging materials and the final management of packaging waste. This method can be a useful tool for selecting the desirable waste management method, but only if the system boundaries are well defined [71].

As with all complex assessment tools, LCA has both its limitations and its strengths [225]. It is a kind of decision-making aid for the design of new products and processes and also serves as a guide for optimising the energy and material consumption of a product or process. The environmental impact of a product's packaging is of great interest as the packaging design, material selection and associated functional requirements are very flexible. The packaging of a product contributes to waste problems, recycling problems and logistics-related emissions. Packaging design takes into account functions such as product preservation, handling, storage, logistics and use. Given the need to consider the sustainability of packaging, many companies are looking to recyclable, lighter, recycled and renewable packaging to make their product more environmentally friendly overall. The literature review shows that only a limited number of LCA studies have been conducted in relation to the manufacturing of pharmaceuticals and pharmaceutical packaging [82,226].

According to the assessment using the eco-design methodology, the relevance of the selected raw materials and their origin can strongly influence the associated environmental impacts, which can also be confirmed quantitatively by the life cycle assessment. A correct methodological adaptation of the concept of "eco-briefing" as a communication tool between environmental engineers and designers, the simplification of the analysis tool used and the application of the life cycle assessment methodology that facilitates environmental analysis are therefore necessary to obtain new formats of packaging materials developed with sustainability in mind [223]. The life cycle assessment makes it possible to compare different systems, taking into account the consumption of resources and the pollutant emissions that can occur during their life cycle, which can include the extraction of raw materials, the production and processing of materials, transport, the utilisation phase and finally the end of the product's life.

The LCA methodology consists of four steps:

1. definition of target and scope,
2. inventory analysis,
3. impact assessment and
4. assessment of improvements.

Many studies used LCA to compare the environmental performance of alternative food packaging systems, either to select the most environmentally sound solution or to assess more specific aspects, such as identifying the most affected categories or the most effective production phases. The environmental impact of the printing industry is significant and is characterised by energy consumption and pollution due to chemical-intensive processes. A great deal of research has focussed on the sustainability of printing. Each printing technique uses different chemicals depending on the type of process shown in *Figure 10.* [5]. LCA can help identify opportunities to improve the environmental performance of products, select relevant indicators, help with strategic planning and apply for an eco-label [227]. In other studies LCA is applied to compare alternative disposal scenarios for the polymers used in packaging (Buccino et al., 2017).

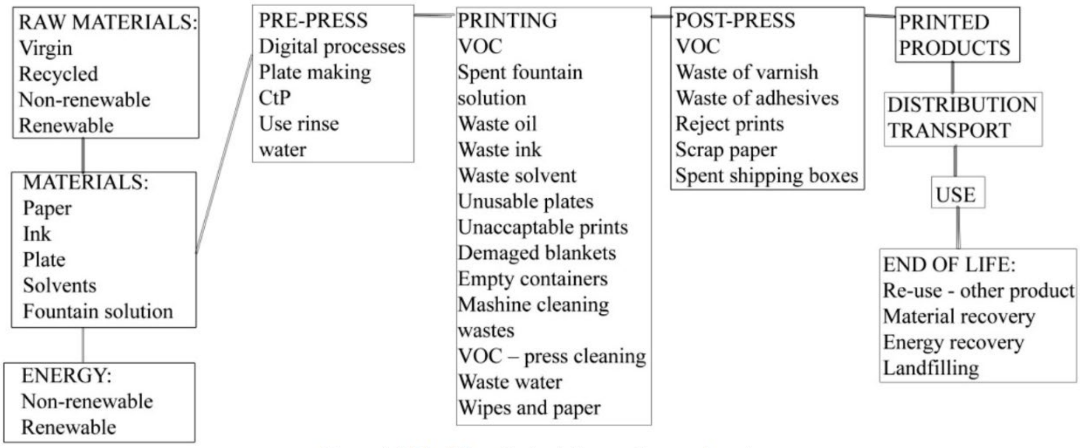

Figure 1 LCA of the offset printing and types of waste

Figure 10. LCA of the offset printing and types of waste [5]

6.4. Eco-design

One of the most effective tools for assessing and reducing the environmental impact of products is eco-design. This method consists of applying environmental criteria to the development of a product and implies a change in the way we look at that product. For a comprehensive analysis, the evaluation of the environmental improvement of the entire life cycle of the product is also taken into account [223]. Eco-design is defined as “the integration of environmental aspects into product design and development with the aim of reducing negative environmental impacts throughout the life cycle of a product”. It must focus on the phases of the product life cycle that have the greatest impact on the environment, so that when the product is redesigned, its environmental impact can be greatly reduced. Integrating eco-design into product development can offer several benefits to industry and public organisations, such as economic benefits, regulatory compliance, stimulating innovation and creativity, improving public image and increasing employee motivation. To be successful and achieve these benefits, eco-design must be integrated into the strategic planning and operational management of the organisation [224]. It must focus its attention on the phases of the product life cycle that have the greatest impact on the environment. If the product is redesigned according to these phases, its impact can be significantly reduced [228,229].

Eco-design is included within the framework of the standard for "Guidelines for incorporating Eco-design" [230].

The eco-design process defined in the standard comprises six steps:

1. Determining the product functions,
2. Environmental assessment of the products,
3. Strategies for improvement,
4. Environmental objectives
5. Product specification and,
6. Technical solutions [231].

6.5. Sustainable Packaging

Sustainable packaging is defined in research of Steenis et al. as packaging that has a comparatively low environmental impact as assessed by a life cycle assessment. Sustainable packaging should be made of non-toxic materials, preferably produced from natural, renewable materials. At the end of its life, it should be recyclable, reusable, biodegradable or compostable [232]. From the consumers point of view, there are different definitions of sustainable packaging. Consumers tend to define sustainable packaging in terms of packaging materials (biodegradability and recyclability) and market appeal (attractive graphic design and good price). When it comes to market appeal, consumer expectations of eco-friendly packaging relate to attractive graphic design, functional performance and price [10]. Packaging design involves a combination of structural, graphic and verbal elements. In this case, the materials are the structural elements and contribute the most to the direct environmental impact. Graphic and verbal elements are informative and both can be used to signal sustainability. Verbal features can be used to communicate sustainability explicitly (e.g. through labelling), while green colouring implicates communicates sustainability [232].

When developing new packaging, the constraints of the packaging industry (scalability of the production process, availability of raw materials, etc.), the regulations and laws for the disposal of packaging at the end of its life (in terms of biodegradability, recyclability, incineration, etc.) and consumer preferences (transparent packaging, environmentally friendly packaging, no additional costs due to packaging) etc. should be taken into account. In the case of

pharmaceutical packaging, the type of packaging has an impact on the emissions generated during the packaging of medicines and their release into the environment.
The increasing use of packaging has resulted in the proportion of paper-based packaging in the waste stream collection system being higher than that of graphic products [169]. When it comes to the use of printing inks in packaging, the amount of printing inks used should be optimised. The printing inks used should also be free of mineral oils and contain the lowest possible proportion of metallic components in the ink formulation. Where possible, a deinkable printing technology should be considered when producing packaging from bleached paper and board. With appropriate collection and sorting, all paper packaging can be recycled. The increased amount of paper collected led to a reduction in the quality of the paper for recycling [202].

7. PACKAGING TODAY

Today, the role of packaging is expanding to include branding, communication, distribution control, poison protection and much more. Packaging has evolved into both a scientific and a technical discipline that has an impact on a product, both within the manufacturing company and with the consumer outside the company. The scientific part of this mix is a broad combination of disciplines, utilised along with drug discovery science as two integral parts of pharmaceutical product development. It has taken on an engineering role, transforming laboratory prototypes and in many cases stable sample packages into a product and package entity that can be safely manufactured, filled, sealed, labelled and distributed. It has also taken on the role of a management tool in the manufacturing process. Packaging is the only scientific and engineering discipline within a pharmaceutical company that touches a product from conception to the complete end of its life or use, including the recycling or disposal of the used packaging [78]. For paper-based packaging products, innovations are being sought that will most likely be laminated with new biopolymers and have new functional barriers capable of replacing multilayer material and smart functionalities, thus replacing at least part of the plastic on the market. All these developments are aimed at achieving a good recycling behaviour of the product, which is indeed one of the most important sustainability aspects of the paper value chain. [169].

Therefore designers are no longer seen just as producers of aesthetic “things”, but are valued for their problem-solving skills and their ability to help companies and organisations develop strategy. In addition, both designers and the clients are expected to consider sustainability issues and take responsibility for the impact of the products and systems. In recent decades, the term sustainability has taken over many environmental and social ideas, economic activities and even a way of life. Dritz defines the term sustainability as *Striving to meet the needs of people, culture and business today in a way that restores the planet and does not compromise our ability to meet the needs of future generations.*

Good design should be sustainable by definition. Responsible choices can simply be added as another criterion for good design, along with form, function, colour and typography. Sustainability should be an integral part of design and define design in terms of its goals and ambitions. We need to consider design as part of sustainability, because sustainability must define our world view, and everything we design should help to communicate that view. Print and paper waste are the first things that come to mind when trying to apply sustainability concepts to the graphic design process. Some commonly applied solutions to these environmental impacts include using recycled paper and vegetable-based inks, as well as creating digital instead of printed materials. Such design that focuses on environmental impact is commonly referred to as eco-design [233].

Designers need to collaborate with other disciplines, understand the interconnectivity of global systems and adopt a "designer-as-social-scientist" approach. This professional shift from "what to design" to "how to design" is radically changing design education (Faerm, 2019). The most important challenge of modern production is therefore the integration of ecological requirements into the process of general management. It is necessary to emphasise the interdisciplinary nature of the postulates of sustainability, which encompasses the fields of economy, ecology and society [97].

For significant improvements in the sustainability of packaging, it is crucial to start with the design of the packaging itself. It is therefore important that we better understand how packaging is designed in practise and the barriers to implementing sustainable design principles. Packaging can be made up of many different layers and materials, each of which needs to be treated in its own way. Recently, more and more attention has been paid to the sustainability of packaging, both in academia and in the media [221]. To increase sustainability, natural resources should be used instead of synthetic ones. Overall, the use of natural resources, environmentally friendly process technologies and end-of-life solutions are required. In terms of sustainable production, the most important parameter is the adhesive, the material used to produce the labels, the printing inks and support on which the label is applied. It is known that multi-layer materials are usually difficult to recover and recycle [202]. When designing a new sustainable graphic product, innovative sustainable solutions should be sought in the area of printing substrates, printing inks, printing process and finishing, as well as options for recycling and waste recovery [227].

7.1. Design

Packaging designers can communicate product benefits through verbal messages, images or stylistic features, implying the right choice of font, image style, use of colours and decorative elements. This requires of them to following market trends to meet the client's design brief and develop appropriate solutions. A designer must consider and balance the different elements while creating a visual hierarchy that allows the viewer to quickly decipher the essential message of the design (e.g. the content of the packaging). These design decisions are often influenced by marketing considerations (e.g. which benefit should be perceived first). Depending on the brief, designers make specific choices for colours, fonts, shapes and configurations and use different types of tools and approaches to solve the often ill-defined problems within a specific timeframe, and each design process is customised [234].

Food packaging serves not only as a container that keeps the contents safe and fresh, but also as a means to communicate the value of the contents and persuade potential consumers to purchase. Consequently, commercial packaging can contain many different messages. In addition to using text, graphic designers can convey a message through the images they use or the style of the package design. The different components must be clearly recognisable to the viewer and should be integrated into a cohesive design. Food packaging usually conveys information whether the product is beneficial to health or not (e.g. nutrition, physical fitness).

The production and consumption of food can additionally have an impact on the physical environment (e.g. biodiversity, use of scarce resources, pollution). Other product benefits include product quality (e.g. flavour, shelf life) or the social and economic aspects of food production (e.g. working conditions, contribution to the community, cultural heritage, small-scale production). The area of health and nutrition claims is strictly regulated in many countries. It is not only about the texts that should or should not be used, but also about the associated visual language [235].

Visual elements included in packaging designs can communicate to consumers on a labelled or connoted level, where labelled level refers to the direct, literal meaning, while the connoted level refers to the more implicit meaning (which may include symbolic aspects). Whether and how people use certain information depends on the perceived relevance of the information, the accessibility of the information and trust in its source. Advantage of using images in a marketing context is that images attract the attention of buyers better. Presenting information in an image appears to be more engaging and vivid than presenting the same information in text form. Processing visual cues seems to require unconscious and involuntary processing, while verbal cues require a higher level of cognitive effort. This is particularly important in large supermarkets, where customers have to find and select a product among many competitors. The textual information on the packaging has a major impact on consumers' expectations of a product. A recent study suggests that interpretative food labelling is perceived as more convincing than reductive labelling. Reductive labels provide information on key nutritional information such as calories, fat, sugar and salt. Interpretive labels help consumers understand whether the contents of the food are good or bad by using specific scores or adding colours. These types of labels can be nutrient-specific or provide a summative average of nutritional value. However, the review also found that consumers are more likely to be persuaded by visual cues on the front of the pack than textual cues.

The phrase a picture is worth a thousand words already indicates that a picture can be a more effective medium for communicating a message than text, as it usually conveys several aspects and more details at the same time. Furthermore, if the designer's intent is unclear, a viewer may be unsure how to interpret a particular image.

One of the benefits of involving designers in the creation of packaging is that they help to develop a design that brings all the requirements together into a coherent and aesthetically pleasing whole. If a designer is unable to create a coherent whole, the packaging design is likely to be perceived as confusing, less compelling or less attractive [235].

7.2. Packaging and Customer Behaviour

Changes in customer behaviour have increased the demand for packaging materials. In order to reduce the overall environmental impact of packaging, the trend towards the development of sustainable packaging has increased over the last decade [202]. Consumers are increasingly concerned about the environmental impact of packaging. There are criteria according to which consumers prioritise the environmentally friendly properties of packaging when choosing food. It has been shown that concern for the environment is positively associated with the intention to purchase environmentally friendly packaging, while price can influence consumers' intention to purchase environmentally friendly packaging [235]. Companies are under pressure not only from consumers but also from governments to use environmentally friendly packaging for their products. Consumers have diverse perceptions of environmentally friendly packaging, but their knowledge is limited and relates more to packaging materials (e.g. biodegradability and recyclability) and market appeal (e.g. an attractive graphic design and a good price). Consumers know little about manufacturing technologies, but still want an environmentally friendly manufacturing process [10].

Eco-friendly packaging has never been a clear concept in the literature. In addition, researchers have used different terms for eco-friendly packaging, such as eco-packaging, ecological packaging, green packaging, sustainable packaging, eco-design, design for the environment and environmentally conscious design and so on leading to confusion in research. In practise, environmentally friendly packaging is often also referred to as sustainable packaging. Many initiatives have been launched to promote the concept of sustainable packaging in the industry. A widely accepted definition of sustainable packaging is provided by the Sustainable Packaging Coalition® (SPC): Sustainable packaging is beneficial, safe and healthy for individuals and communities throughout its life cycle; it meets market criteria for performance and cost; it is sourced, manufactured, transported and recycled using renewable energy; it maximises the use of renewable or recycled source materials; they are manufactured using clean production technologies and best practises; they are made from materials that are healthy in all likely end-of-life scenarios; they are physically designed to optimise materials and energy; and they are effectively recovered and recycled in biological and/or industrial cradle-to-cradle cycles [10].

Packaging can be functionalised to prevent food loss and waste by extending the expiry date and adjusting the packaging size, but there may also be additional environmental impacts from changes in food and packaging production. Previous studies on these topics have analysed the additional impacts of packaging production. However, the impact of packaging functionalisation has yet to be linked to food production and consumer behaviour [236]. Sustainable packaging is expected to protect the product and communicate its features, encompassing the reuse of materials and the reduction of waste throughout the life cycle of a package from production to consumption, disposal and end-of-life [10].

In the study by Yokokawa et al. products with functionalised packaging that allow a longer expiry date or a smaller portion size were compared with their base products. The results showed that the packaging-related changes increased the global warming potential (GWP) of food production more than other processes. Thus, the changes in food production weakened the effectiveness of packaging functionalisation to reduce GWP. In addition, the analysis of consumer behaviour scenarios showed that consumer perception of the expiry date has a decisive influence on the effectiveness of packaging functionalisation. When consumers discarded food after the expiry date, provided they consumed it in small quantities, packaging functionalisation reduced food loss and waste (FLW), identifying suitable combinations of packaging functionalisation and consumer behaviour to effectively decrease. The analysis can help packaging designers understand the effectiveness of their product life cycle decisions in reducing FLW and environmental impacts [236]. The study by Escursell et al. showed that 71% of e-customers would shop online again if they received high-quality packaging. It is therefore important to improve the consumer experience through functional and aesthetic presentation of packaging and preservation of their brand image. They should strive to control the direct and indirect environmental impact of their packaging through green messages [237].

Packaging materials should not only be environmentally friendly, they should also be able to easily handle the "last mile", which is currently the biggest obstacle to the expansion of e-commerce. Companies are struggling to deliver lighter packages as quickly as possible, but of course this comes at a price. IKEA, for example, charges a high price for home delivery of its products, but it is its customers' choice to pay this price [238]. Standard stores are increasingly using paper bags instead of corrugated board packaging for home [239,240], saving 80 % energy.

8. CARBOARD PRODUCTION DESIGN AND OPTIMIZATION

Controlling and minimizing packaging materials and packaging waste is an important part of all sustainability initiatives today. Despite the global recognition of the importance of waste reduction in the packaging of materials and products, little research has been done to understand how packaging solutions are developed within supply chains and the impact these solutions have on the rest of the supply chain. There is a different relationship between packaging and sustainability. Packaging is designed to be disposed of once it has fulfilled its basic functions, so the energy required to produce packaging is often not commensurate with its relatively short lifespan. Once the product has been delivered, the packaging itself has fulfilled its function and its life cycle has come to an end. The importance of reducing packaging waste has gained renewed importance and awareness due to the COVID-19 pandemic, which has led to an increase in demand for single-use products and the use of packaging. Companies and organizations are setting goals and committing to adopting sustainable practices and investing in packaging optimization to meet consumer demands and further improve the efficiency and effectiveness of packaging [241].

Improving the efficiency of packaging is an important strategic goal that can lead to significantly improved environmental performance. Packaging design can offer significant cost and environmental benefits and improve product reliability. Studies have shown that the decisions around packaging design are more complicated than simply selecting the most optimal environmental solution [242]. Recent work related to the development of new phenomenological models has focused on design. In the face of increasing global competition, it is necessary to design products that are able to fulfil needs in the most effective way. [243]. The literature on packaging design of packaging materials is largely concerned with aesthetics, function and the role of packaging in influencing consumer buying behavior. However, packaging materials have a much broader purpose than just consumer appeal; they also have an environmental dimension. [242]. Challenges related to the quality of packaging must be overcome. To determine the right optimization model, it is important to understand both the process and the material (paper) in order to improve pulp quality.

Optimization is a biological instinct that is evident in the evolution of species. Humans are constantly trying to improve their lives and the systems around them, and animals do the same (e.g. birds optimize the shape of their wings in real time, and dogs have been shown to find optimal flight paths). Even more broadly, many laws of physics relate to optimization, such as the principle of minimum energy. As Leonhard Euler once wrote, “nothing at all takes place in the universe in which some rule of maximum or minimum does not appear.” It can therefore be said that optimization has permeated all areas of human endeavor. Although optimization has been practiced in one form or another since the early prehistoric era, this field has seen increasing growth in the last five decades. It has therefore become essential to plan, design, operate and manage resources and assets optimally [244].

The terms “optimize, optimization, optimal, optimum” are often used in a very loose sense, without necessarily referring to the use of specific optimization techniques. Every time a product is created or designed to satisfy human needs, the designer tries to find the best solution for the task at hand and therefore carries out optimization [245].

The term optimization is often used in the sense of “improvement”, but mathematically it is a much more precise concept: finding the best possible solution by changing variables that can be controlled, often subject to constraints. Optimization has great appeal because it is applicable in all areas and because humans have a desire to improve things. Any problem where a decision needs to be made can be represented as an optimization problem. Although some simple optimization problems can be solved analytically, most practical problems of interest are too complex to be solved in this way.

In general, design optimization is widely used in various engineering disciplines, such as the design of airfoils in aerospace engineering, process control in chemical engineering, the design of structures in civil engineering, the design of circuits in electrical engineering, and the design of mechanisms in mechanical engineering. Most technical systems rarely work in isolation and are connected to other systems, so design is a multidisciplinary activity [244]. Therefore, this process involves teams of experts and multiple stages with many iterative loops that can be nested. Design optimization can replace an iterative design process to speed up the design cycle and achieve better results [246]. A simplified version of the conventional design process compared to the design optimization process is shown in *Figure 11*.

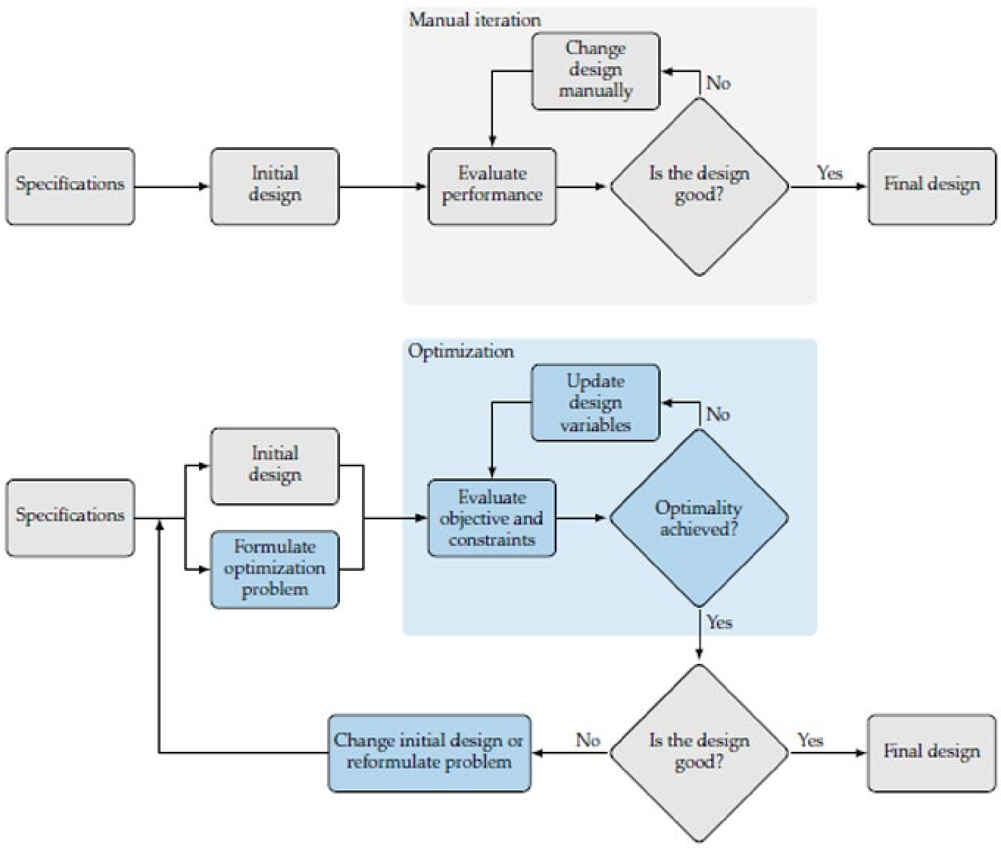

Figure 11. The conventional engineering design versus design optimization process [246].

Optimizing sustainable practices for the design and recycling of packaging requires knowledge of the design phase, the design variables and their minimum and maximum limits (independent variables), the constraints, the measurement of design performance (dependent variables), the design parameters and the relationships between the independent and dependent variables (i.e. a design evaluation model). The design variables depend on the level of product definition available at the various stages of design optimization. The design stages can range from conceptual or preliminary design and configuration to detailed design. Design variables are expressed in either quantitative or qualitative terms. For many design optimization problems, it is easy to measure design variables such as length, weight and temperature. They are called quantitative design variables. However, in real-world design problems, variables such as esthetics and manufacturability are difficult to measure and are referred to as qualitative variables. The type of model development depends on the types of variables [243].

Optimizing the use of adhesive and laminate in packaging involves several considerations aimed at reducing waste, ensuring product protection and minimizing environmental impact. Here are some strategies to achieve this:

1. Material selection – Choose packaging materials that require minimal adhesive or laminate to maintain structural integrity. Corrugated boxes, for example, typically require less adhesive than plastic packaging.
2. Design efficiency – Optimize packaging design to minimize the need for excessive glue or lamination. Use structural techniques such as tabs, slots and interlocking features to create strong packaging without relying heavily on adhesive.
3. Alternatives to lamination – explore alternative methods to improve the durability of packaging without excessive use of laminate. This could include surface treatments such as water-based coatings or varnishes that provide protection against moisture and abrasion without the need for additional layers of lamination.
4. Precision application - Use precise application techniques for applying glue or laminate to reduce waste and ensure optimal bonding. Automated dispensing systems can help ensure consistent application with minimal excess.
5. Recyclable and biodegradable materials - Whenever possible, favor the use of recyclable and biodegradable packaging materials. These materials typically require less adhesive or lamination and have a lower impact on the environment throughout their life cycle.
6. Testing and quality control - Conduct thorough testing to ensure that packaging designs and materials meet durability and protection requirements with a minimum of adhesive or lamination. Implement quality control measures to identify and correct any problems early in the production process.
7. Collaborate with suppliers - Work closely with packaging material suppliers to find innovative solutions to reduce the use of adhesives and laminates while maintaining packaging performance. Suppliers may have access to new materials or technologies that can help optimize packaging designs.
8. Consumer education - Educate consumers on the importance of responsible packaging disposal and recycling. Encourage them to properly separate packaging materials to facilitate recycling and reduce environmental impact.

By implementing these strategies, companies can optimize the use of adhesives and laminates in packaging to achieve a balance between product protection, sustainability and cost efficiency.

9. METHODOLOGY

The experimental part of this work is divided into several key sections. Some of the results previously published in peer-reviewed journals are presented in chapters 1-8, while results that have not yet been published are presented in chapters 9-10. In this way, a comprehensive overview of research and results is provided. The published works represent key findings that are the basis for further research into the optimization of the cardboard packaging product, i.e. the creation of a prototype of a viable pharmaceutical packaging product. The integration of these findings into the doctoral thesis enables a comprehensive understanding of the research process and results, providing continuity between the experimental part and the final conclusions.

The research was conducted according to the proposed plan including the following:

1. Recycling of six samples series with three different flotation methods;
2. Production of laboratory sheets of paper and sheets of foam;
3. Image analysis and determination of the optical properties of recycled laboratory sheets and foam sheets;
4. Analysis of metal traces in samples using the ICP-MS method;
5. Proposal for the optimization of the manufacturing process of laminated cardboard pharmaceutical packaging product;
6. Conceptual Solution of a Sustainable Pharmaceutical Product.

9.1. Materials

For the purpose of this research the printing substrate, the printed quire and the printed packaging were analyzed. The printing substrate was GC2 cardboard, to which an acrylic polymer- based dispersion was applied. The cellulose and other components of the cardboard printing substrate meet the conditions for food and pharmaceutical products, which means that it does not contain increased concentrations of metals and other harmful components. This is crucial for analyzing the concentration of metals, which is one of the subjects of this research. The printing substrate is also a sustainable product, as it consists of 60% virgin fibers and 30% high-quality post-industrial fibers. The front side of the printing substrate is coated with a three-layer pigment varnish, while the back side is only coated once.

The laminated printing surface is produced with a dispersion based on acrylic poly-mers (biaxially oriented polyethylene terephthalate), also known as metallized BoPET film on the same GC2 cardboard. These self-crosslinking acrylics are free of plasticizers and alkylphenol ethoxylates (APEO). The APEO compounds can affect the environment, aquatic organisms and humans. The data show that short-chain compounds have a much lower impact than long-chain compounds [247]. The function of the dispersion is to apply a biaxially oriented polyethylene terephthalate film (BOPET metallized) to obtain a laminated packaging material. The film complies with the requirements of the repealed Directive 20/590/EEC.

The prints were prepared with UV offset inks produced by Sun Chemical® Europe. The printing process started with white offset printing ink, continued with CMYK separation and a dark purple-blue pantone color was used at the end of the printing process for the text on the packaging. For rub resistance, the prints were varnished with a UV-cured varnish, which is a highly reactive photopolymerizable acrylate system, VOC free, with reduced odor and optimal wetting properties. The UV-cured varnish mentioned is VP 1038 high gloss, VergamGH (marked L2). For assembling the packaging, an adhesive was applied to the edges in compliance with the European framework directive 89/109/EEC, specific rules for adhesives in food applications and regulation (EC) of the European Parliament and of the Council on materials and articles intended to come into contact with food and repealing Directives 80/590/EEC, and Commission Regulation (EU) No. 10/2011 on plastic materials and articles intended to come into contact with food [248,249].

9.2. Samples

The samples used in this research were selected based on specific criteria, main being the presence of lamination and the stage of the cardboard packaging manufacturing process. By applying these criteria, it is ensured that the samples accurately reflect the characteristics of cardboard pharmaceutical packaging in the recycling process. Stage of packaging production in the experiment for non-laminated and laminated samples is shown in *Figure 12*. Samples used for recycling and their labels are shown in *Table 1*. The samples are as follows:

1. Printed packaging – coated substrate with Ag-polyethylene + ink based on renewable raw materials + printing with UV curing ink + UV cured varnish + product assembly adhesive.

2. Printed quire – coated substrate with Ag-polyethylene + ink based on renewable raw materials + printing with UV curing ink + UV cured varnish.
3. Printing substrate – coated substrate with Ag-polyethylene.
4. Printed packaging – uncoated substrate + ink based on renewable raw materials + printing with UV curing ink + UV cured varnish + product assembly adhesive.
5. Printed quire – uncoated substrate + ink based on renewable raw materials + printing with UV curing ink + UV cured varnish.
6. Printing substrate – uncoated substrate.

Table 1. Samples used for recycling and their labels

Type of sample	Label	Sample	Label
Laminated	L	Printed packaging	P
		Printed quire	Q
		Printing substrate	S
Non-laminated	N	Printed packaging	P
		Printed quire	Q
		Printing substrate	S

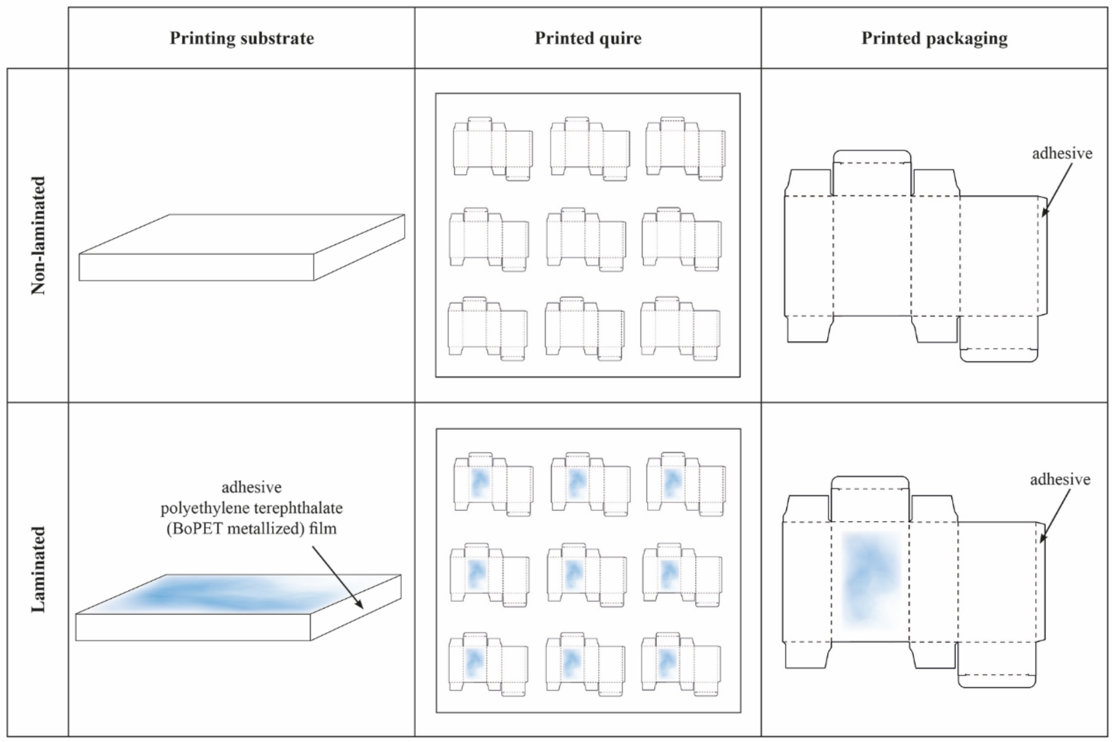

Figure 12. Stage of packaging production in the experiment

9.3. Equipment and Machinery

Offset printing machine

Printing samples were made with a standard printing form on a five-color offset machine, Man Roland 705, a well-known German printing press manufacturer. This press offers reliable, high-volume production with superior color quality. It is suitable for printing packaging materials that require high-quality color reproduction and precise registration. It is used on a variety of substrates, including different types of paper, cardboard, and possibly other materials used in specialty printing.

Printing form contained different printing elements: standard CMYK step wedge in the range from 10-100% tone value, standard ISO illustration and the standard wedge with 378 patches for production of ICC profiles.

Disintegrators

Disintegrators are machines used in the papermaking industry. They play a crucial role in paper recycling, as they help to efficiently break down (old) paper materials into a form that can be reused to produce new paper products, reducing the need for virgin pulp from trees and minimizing waste. As a result, the formed suspension of cellulose fibers in water is called paper pulp. The obtained pulp can then be processed and used to create new paper products.
The disintegrator typically consists of rotating blades or hammers that shred the paper material into small fibers or pieces. These fibers are then mixed with water to form a slurry, which undergoes further processing in the papermaking process, such as refining, screening, and drying, to produce new paper sheets. In this work, an Enrico Toniolo disintegrator was used.
Disinegrator manufacturer Enrico Toniolo
Reference standards:
uni en iso 5263-1: 2005 – chemical pastes;
uni en iso 5263-2: 2005 – mechanical pastes
Tappi t 205; scan c18 / m^2; paptac c.6
Built with high quality materials, balanced hinged head, digital revolution counter and display, speed of revolution according to iso and tappi regulations.
– acrylic glass disintegrator pot
– power supply 230v / 50 hz ac
– net weight 40 kg

Homogenizer

Homogenization with a pulping homogenizer is a piece of equipment used to further refine the pulp produced by the disintegrator, which helps ensure uniformity and consistency in the resulting pulp by preventing the settling of cellulose fibers during the production of laboratory sheets according to ISO standards. It typically consists of a series of rotating blades or disks that further refine and mix the pulp, breaking down any remaining large fibers or clumps and ensuring that the pulp has a consistent texture and composition. This helps improve the quality of the pulp and ensures that it meets the desired specifications for use in papermaking. Overall, the pulping homogenizer plays an important role by helping to produce high-quality pulp that can be used to manufacture a wide range of paper products.
After the homogenization process, the paper suspension is transferred to the flotation cell, where the flotation process takes place.

Flotation cell

Flotation cell is a device used in mineral processing to separate valuable cellulose fiber from impurities through the process of flotation. In flotation, air bubbles are introduced into the pulp. The hydrophobic particles (those that repel water) attach to the air bubbles, rise to the surface, and form a froth layer, which is then skimmed off. The hydrophilic particles (those that attract water) remain in the water and are discharged as tailings.To increase the efficiency of deinking flotation, certain chemicals are added to increase the hydrophobicity of the particles.
Flotation cells come in various designs and sizes, but they typically consist of a tank or vessel where the flotation process occurs, along with mechanisms to introduce air bubbles and agitate the mixture to enhance particle attachment to the bubbles. The froth containing the valuable minerals is usually collected from the top of the cell, while the tailings are removed from the bottom.

Sheet Former Rapid Koethen

Laboratory sheets before and after flotation were made on the Rapid Kothen Sheet Former Machine, an automatic paper-making device. This device is used to produce laboratory sheets for measurement purposes. The cylinder is filled with water up to a level of 4 litres, and the suspension is added to the device. When the suspension and water are evenly mixed, the device releases excess water through the bottom of the tank, leaving fibres on the wire mesh, resulting in a wet-formed laboratory sheet. A highly absorbent paper is placed on this sheet, which is then manually separated from the wire mesh and placed in a drying device for 8 minutes.
The Sheet Former is built upon a warp-resistant stainless steel frame. This robust base holds the pump, hot water bath, suction chamber, etc. On the left-hand side of the working table you can find the sheet forming column. The dryer is located on the right-hand side. There are only a few important control switches on the working table to ensure the workspace for the transferring process between the sheet former and the dryer. The sheet forming process is controlled by a Simatic control system. The temperature and the vacuum of the drying process are adjusted on the touchscreen.

Spectrophotometer

Spectrophotometer measures the optical characteristics of paper pulp and paper. The device operates by breaking down white light into individual wavelengths using a monochromator (prism or optical grating). This allows for the measurement of changes in reflection, radiation, or transmission at intervals along the wavelengths of the visible spectrum. In this work, we are using the Technidyne Color Touch 2 spectrophotometer.

Measurements include:

- ISO Brightness
- ISO Color and Color Difference, including Whiteness, Tint, Yellowness and Metamerisam, ISO Fluorescence
- ISO Opacity, Scattering and Absorption
- TAPPI Effective Residual Ink Concentration (ERIC 960)

Image analysis

The number and surface area of residual impurities particles were determined using image analysis software, Apogee's Spec*Scan 2000, with a sample digitization scanner. The particles were divided into 26 size classes [250]. Spec*Scan 2000/2001 is a grayscale image analyzer for pulp, paper, and paperboard. Images are obtained using a desktop scanner and analyzed for specks, dirt, residual ink, pulp shives, printed patterns, and other objects that optically contrast with the paper substrate by size and reflectivity. The software also measures their size, location, grayscale, and shape and generates reports. The measurements, of both size and reflectivity, are following TAPPI T-437 which states the estimated equivalent black area of a gray or colored speck is smaller than its actual area in inverse proportion to the intensity of its color contrast with its background. It uses a simple statistical method to quantify differences in formation quality by analyzing the frequency distribution of the grayscale value of each pixel in the specimen sample image.

Spec*Scan contributes to optimizing pulp screening and bleaching systems, maintaining cost control for raw materials and pulp mixing. It provides accurate, reproducible dirt content quality control and controls stickies content in recycled fiber.

9.4. The Workflow of the Experimental Part of the Research

The first part of the study involves determining the efficiency of print recycling and characteristics of recycled fibres. Samples are converted into pulp through disintegration processes ISO 5269-2:2004 [251] and particle separation occurs using three different methods for all six-sample series:

1. INGEDE 11, according to ISO 5263-2:2004 [252],
2. Flotation deinking method with adhesive particle separation,
3. Adhesive particle separation method.

Recycled paper samples are produced on a standard laboratory sheet former Rapid-Köthen device. Optical measurements are conducted on laboratory paper sheet samples according to standard methods:

- Diffuse blue reflectance factor according to ISO 2470-1: 2016 [253]
- Residual ink concentration, ERIC according to TAPPI T 567: 2009, ISO 22754: 2008 [254,255]
- Paper and board colour determination, ISO 5631-3: 2015 [256]

Using Spec*Scan Apogee System image analyzis software with a sample digitization scanner, the number and area of residual impurity particles, divided into 26 size classes ISO 13322-1, 2014 [250], are determined.

The second part analyzes traces of metal in paper samples using Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Metal concentration is quantitatively measured. The study determines the metal distribution in different phases of the recycling process in laboratory sheets and foam samples. Most analyzed metals are found in the paper pulp or samples as a result of printing ink composition.

The recycled paper sample was cut into small pieces and then weighed to approximately 100 mg. To extract the metals from the paper, 5 ml of hydrochloric acid and nitric acid (1:3 ratio) were added to the sample (J.T. Baker, p.a. purity). After dilution, the analy-zis was carried out so it is the sample was filtered with a syringe filter and then diluted ten times. Elemental analyzis (Ag, Co, Cr, Cu, Fe, Mn, Ni, Ti, V, Zn analyzis) was performed by inductively coupled plasma mass spectrophotometry (ICP-MS PerkinElmer SCIEX ™ ELAN® DRC-e, Concord, ON, Canada), which applies continuous scattering.

The third part of the study involves designing a sustainable design for pharmaceutical cardboard packaging products based on the results of previous research. The conceptual design will be created using the InDesign software.

After converting the sample into cellulose pulp (disintegration process) according to ISO 5263-2:2004, the separation of impurity particles was performed in three ways. One of the methods, Procedure P1, was performed without chemicals. This procedure is intended to increase the sustainability of the deinking process and improve the production process of the raw material or paper. The following method, Procedures P2, is a combination of the standard INGEDE Method 11 and Process 1 [257,258]. The standard INGEDE Method 11 of deinking flotation was used as a test framework against which laboratory sheets of paper obtained by other methods were compared. The flow diagram of the process of the procedures in the production laboratory sheet is shown in *Figure 13*. The handsheets are made according to the standard INGEDE 1 procedures and ISO 5269- 2:2004 [251] standards, in all the phases of the P1, P2 and P3. Procedures using the RapidKöthen sheet former.

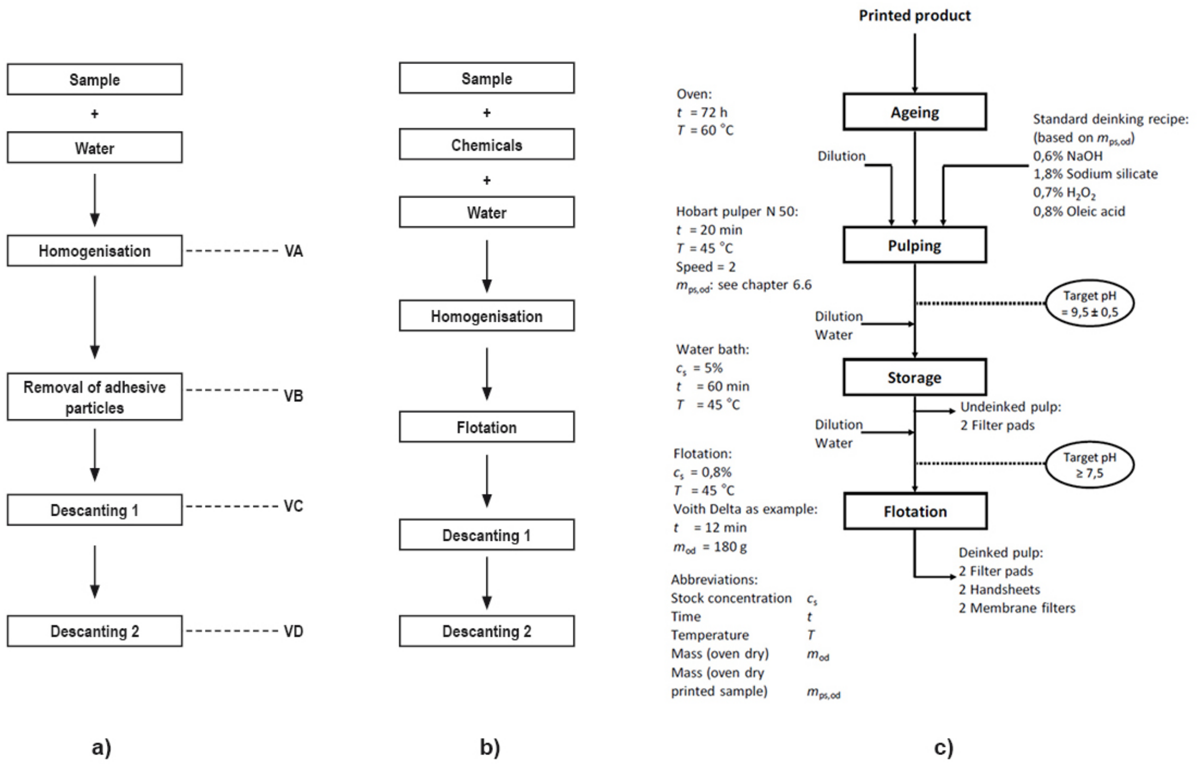

Figure 13. The flow diagram of the process of the procedures in the production laboratory sheet.

Deinkability was evaluated by calculating the flotation yield, the increase of brightness and whiteness, and colour properties of the pulp, by determining residualink area, as well as ash content elimination. The following methods were used for measuring the optical characteristics of laboratory handsheets: the diffuse blue reflectance factor according to ISO 2470-1:2016 [253] effective residual ink concentration, ERIC according to TAPPI T 567:2009 [254], ISO 22754:2008 [255] and color determination for paper and board, ISO 5631-3:2015 [256]. Image analysis was used to count the detected residual impurity particles and area. Spec*Scan Apogee System image analysis software according to ISO 13322-1, 2014 [250] was used, which includes a scanner to digitalize images. The values on the device were as follows: threshold value (100), white level (75) and black level (65) were chosen after comparing the computer images to the handsheets.

The samples were disintegrated into cellulose pulp, according to ISO 5263-2:2004 [259]. The standard flotation deinking method INGEDE 11 [257] was used for the separation of the ink particles from the cellulose pulp. The handsheets were produced according to the INGEDE 1 procedure (Ingede 1, 2007) and ISO 5269- 2:2004 standards. The standard handsheets for this study were produced using a Rapid-Köthen sheet former manufactured by Frank – PTI according to the ISO 5269 standard method.
Some of the optical properties of the laboratory handsheets were measured according to the standards presented in *Table 2*.

Spec*Scan Apogee System image analysis software was used to determine the count and surface area of the remaining dirt particles. A scanner digitised the images with the following settings: Threshold (100), White Level (75) and Black Level (65) [260].
With the spectrophotometer Technidyne Color Touch 2 was analysed the effective residual ink concentration (ERIC number) and CIE L*, a*, b* chromatic coefficients on laboratory handsheets before and after deinking flotation. The effective residual ink concentration on samples is measured according to standard methods ISO 22754:2008 [255] and TAPPI T 567: 2009. The standards according to which the chromatic coefficients were measured are shown in *Table 2*.

Table 2. Optical measurements performed on the laboratory handsheets and the standard methods employed

Optical properties	**Standard**
Diffuse blue reflectance factor	ISO 2470-1:2016

Effective residual ink concentration, ERIC	TAPPI T 567: 2009, ISO 22754:2008
Determination of color by diffuse reflectance	ISO 5631-3: 2015
Image analysis	ISO 13322: 2014

In the recycling process alkali chemical deinking was used. By applying the method of releasing adhesive particles, no surface-active substances are used, contributing to process sustainability. Sodium hydroxide is added to all processes, giving the pulp an alkaline character (pH = 9.0 – 11.0). This pH value favors the reactions of saponification and/or hydrolysis of resins from printing inks as well as the swelling of cellulose fibers, which makes them more flexible (better separation of the ink particles). H_2O_2 is used to lighten and prevent darkening of the pulp. Therefore, in the procedures with H_2O_2 and Na_2SiO_3, a silicate is included to prevent the decomposition of H_2O_2. The positive proper-ties of $Na_2SiO_3$are also evident in the reduction of surface tension, the effect on particle dispersion, and the prevention of the binding of impurities. When determining metals in paper laboratory sheets and metal extraction foam, the following reagents are used: a mixture of hydrochloric and nitric acids in a 1:3 ratio (J.T. Baker, p. a. purity).

The next step of the study was to determine the metal amounts in the laboratory paper respectively in cellulose pulp. The method used for the analysis is inductively coupled plasma mass spectrometry (ICP-MS). The ICP-MS system measures the concentration of elements quantitatively and gives the total amount of each element of interest. The process can be divided into four stages: Sample feed, ICP torch, interface and MS.

The recycled paper sample was cut into small pieces and then weighed to approximately 100 mg. To extract the metals from the paper, 5 ml of hydrochloric acid and nitric acid (1:3 ratio) were added to the sample (J.T. Baker, p.a. purity). After dilution, the analysis was carried out so it is the sample was filtered with a syringe filter and then diluted ten times. Elemental analysis (Ag, Co, Cr, Cu, Fe, Mn, Ni, Ti, V, Zn analysis) was performed by inductively coupled plasma mass spectrophotometry (ICP-MS PerkinElmer SCIEX ™ ELAN® DRC-e, Concord, ON, Canada), which applies continuous scattering. ICP-MS is a tool for analyzing trace metals in environmental samples. In this method, the sample is atomized to generate atomic and small polyatomic ions, which are then detected. The working conditions of the device are listed in

Table 3. It is possible to detect metals and various non-metals in liquid samples at very low concentrations.

Table 3. The working conditions of ICP-MS PerkinElmer

Spray gas flow rate	0, 85 L/min
Auxiliary gas flow rate	1, 2 L/min
Plasma flow rate	14 L/min;
Lens Voltage	8.5 V; ICP RF
Power supply	1, 100 W; CeO/Ce = 0.016; Ba ++/Ba + = 0.015

The ICP-MS calibration was carried out using certified standards. Internal standards are used to compensate for possible measurement deviations. A large number of elements can be detected with an ICP-MS. More than 70 elements can be measured simultaneously in a single analysis. It can measure virtually every naturally occurring element plus many non-natural "radiogenic" isotopes such as technetium, neptunium, plutonium, and americium. The only elements that ICP-MS can't measure are H and He (which are below the mass range of the mass spectrometer), Ar, N, and O (which are present at high levels from the plasma and air), and F and Ne (which can't be ionized in an argon plasma) [261]. The advantages of using a plasma over other ionization methods, such as flame ionization, are that the ionization takes place in a chemically inert environment, which prevents the formation of oxides, and the ionization is more complete. In addition, the temperature profile of the torch is relatively uniform, which reduces self-absorption effects. Linear calibration curves over several orders of magnitude are observed for ionization processes.

For mass spectrometry, the generation of particles in the submicron range with efficient particle transport to the ICP plays a decisive role [262]. The development and use of the plasma torch as an excitation and ionization source in spectrometry has brought about an important development in analytical elemental analysis. Nowadays, ICP-MS is an essential analytical technique in various fields. This technique requires simple spectra, adequate spectral resolution and low detection limits for nearly all the elements it can measure. It can detect many elements at levels below 0.1 part per trillion (ppt). It can also measure elements at concentrations up to 100s or even 1000s of parts per million (ppm). All metals whose presence was analyzed in this work have a detection limit of up to 10 ppt [261]. For this reason, a mass spectrometer was used as a detector and a high-pressure plasma as an ion source [263]. The method ICP-MS was used for multi-element analysis for the quantitative determination of silver (Ag), cobalt (Co), chromium (Cr), nickel (Ni), iron (Fe), manganese (Mn), titanium (Ti), vanadium (V) and zinc (Zn). The mass fraction of all metals is determined according to formulas 1 and 2.

Species mass fraction in the mixture = $Wi = \frac{\text{Mass of specie 'i' in mixture (mg)}}{\text{Total mass of mixture (kg)}}$ 1)

$\sum_{n=1}^{I=n} Wi = 1$

10. CONCEPTUAL SOLUTION OF A SUSTAINABLE PHARMACEUTICAL PRODUCT

The initial design and its specifications were defined and the design problem was formulated. The task required expertise in both the subject area and numerical optimization. Therefore, research was carried out in the field of paper sustainability, packaging design and chemical process engineering. Starting from the conceptual design and optimization of a material using different mathematical models, a new packaging was designed and an optimization was proposed, which is the main contribution of this thesis.

The optimization had to be carried out in order to save material and create more sustainable packaging. Before the design optimization process, the requirements and specifications were defined. We developed a methodology to optimize the design of intelligent pharmaceutical packaging, taking into account product safety and quality. To do this, we conducted in-depth research on pharmaceutical packaging and an analysis of current design. In contrast to most research, we proposed a design concept for a new, improved pharmaceutical packaging. As an innovation, a numerical calculation model was developed to calculate the optimization of the use of adhesive and laminate. The methodology consists of a system of methods for design and optimization with different mathematical models to estimate material savings and thus reduce the environmental impact. The proposed methodology will reduce experimental work by identifying configurations that could be optimal.

Optimization of the packaging is achieved by obtaining the best visual and protective results for the printed, i.e. front/marked side of the packaging. For this purpose, it is important to:

- Choose suitable materials that enhance the appearance and protection of the printed side. Common options are glossy, matt or soft touch films. Pay attention to which gloss or matt finish suits the design;
- Prepare the print to ensure high quality and resolution of the design on the front, resulting in a clear and vivid print, which is particularly important for visual impact;
- High quality printing is essential for front side lamination with accurate color reproduction and sharp details;
- During the lamination process, special care should be taken to ensure that the front side is aligned with the laminating film to avoid misalignment;
- It is necessary to perform quality control to detect defects on the laminated material. The quality control includes a visual inspection, a touch test and other quality checks.

In order to obtain a more environmentally friendly packaging, the profit rate function was optimized based on an economic analysis. A typical roll of metal foil is 995 mm x 3.000 m in size. With 10.000 sheets of 710 x 1.010 mm, 7.770,95 m^2 of foil are used at a price of 0,11 €/m^2, which amounts to 854,80 € in this calculation. It is known from printing practice that an average of 95 kg of laminating glue (Planatol KF 591) is used to laminate 10.000 sheets in 710 x 1010 mm format. The consumption of glue for joining the boxes is minimal. The glue is applied to the sheet, then a metal foil (K12 Pet Met Silver) is placed on top of the glue. The foil is applied at a width of 71 mm, with some excess. The typical price for metal foil is 0,11 €/m^2. *Figure 14.* shows packaging printed substrate with the dimensions.

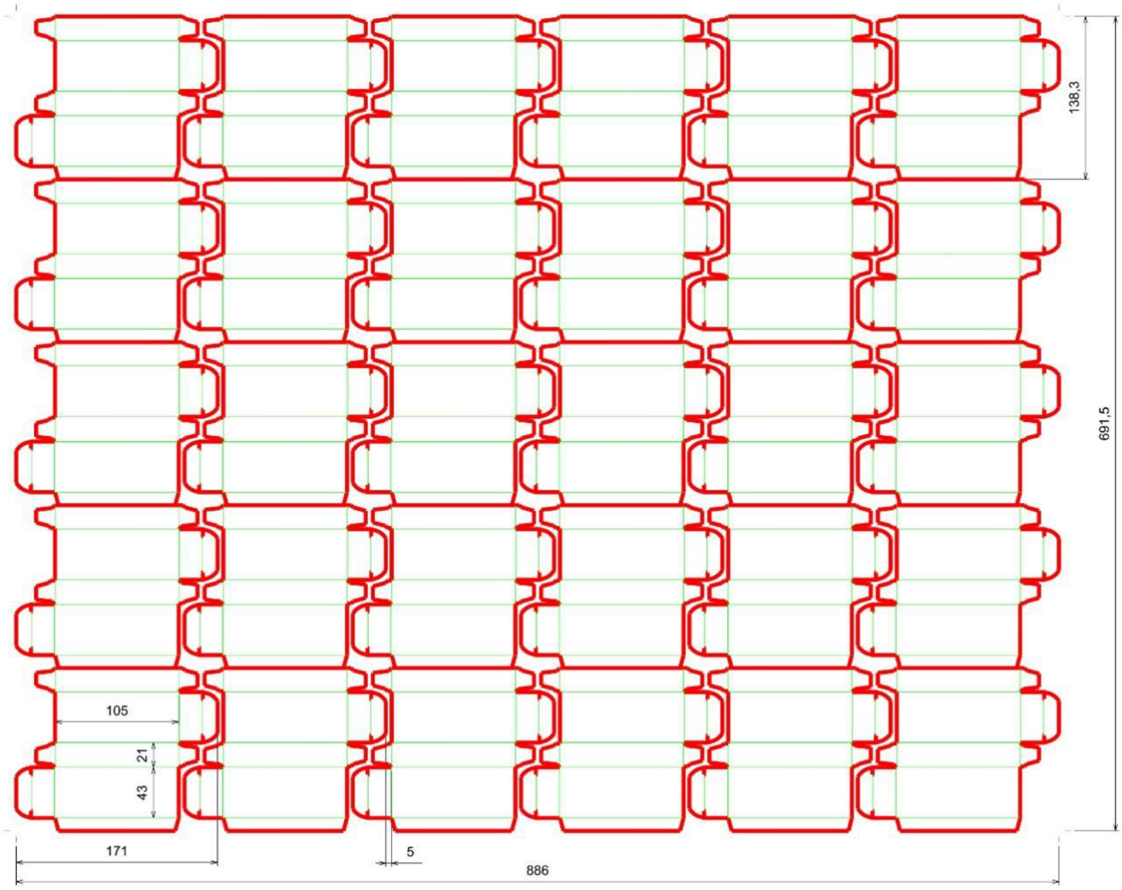

Figure 14. Printed substrate packaging with dimensions

To minimize waste, it is suggested that only the front of the packaging is laminated. Accordingly, it can be estimated how much material is saved for a typical box of pharmaceutical products if the lamination of the other sides of the packaging is omitted. In this way, the production cycle can be optimized and ultimately the environmental impact during production can be reduced. The environment is also protected when used boxes are disposed of, and the recycling of packaging is facilitated and accelerated.

A packaging with the typical dimensions of 105 (width) x 43 (height) x 21 (depth) mm was taken as an example of the savings. The calculation of the relative savings achieved and the savings for packaging with the specified dimensions can be carried out as follows:

First, the consumption per unit area is calculated:
consumption per unit = 95 kg / (71 x 11 cm) / 10 000 quire = 0,012164 g / cm^2 = 1,2164 g / dm^2
2)

On this basis, it is possible to calculate how much savings can be achieved per piece of packaging:

Savings (kg) = 2 x (a x b) x 2 x (a x c) x 2 x (b x c) x consumption per unit – 2 x (a x c) x 2 x (b x c) x consumption per unit 3)

The savings can also be expressed as a relative amount in percentages:

Relative savings (%) = {[2 x (a x b) x 2 x (a x c) x 2 x (b x c)] – [a x b] [2 x (a x b) x 2 x (a x c) x 2 x (b x c)]} / x 100 4)

where *a* is the width of the box, *b* is the height of the box and *c* is the depth of the box.

The savings on metal foils can be calculated in a similar way:

Consumption per unit = O,11 €/m^2

On this basis, it is possible to calculate how much savings can be achieved per piece of packaging:

Savings (€) = 2 x (a x b) x 2 x (a x c) x 2 x (b x c) x 0,11 €/m^2 5)

As mentioned afore, three different design concepts for the packaging are proposed and considered. The design cycle started with an initial design, which was based on past designs adding a new value to it (obtained by the study). Looking from structure perspective, boxes can be made a) by minimizing lamination b) without adhesive and c) combination of the two.

We propose a box where only one side is laminated, with perforations that allow easy removal of the laminated part, as shown in Figure. This feature allows for easier sorting of waste and therefore recycling, while maintaining packaging main role of the packaging. The construction of the box remains intact and retains its recognizable shape. In this way, it does not confuse the customer. The attractiveness is maintained by laminating the side visible to the customer, keeping the product influential and competitive in the market. The target audience is environmentally conscious customers who are likely to opt for eco-friendly packaging as they have the option to separate and recycle the packaging. In addition, the consumer can easily recycle the packaging themselves. *Figure 14.* shows an example of optimized sustainable pharmaceutical packaging.

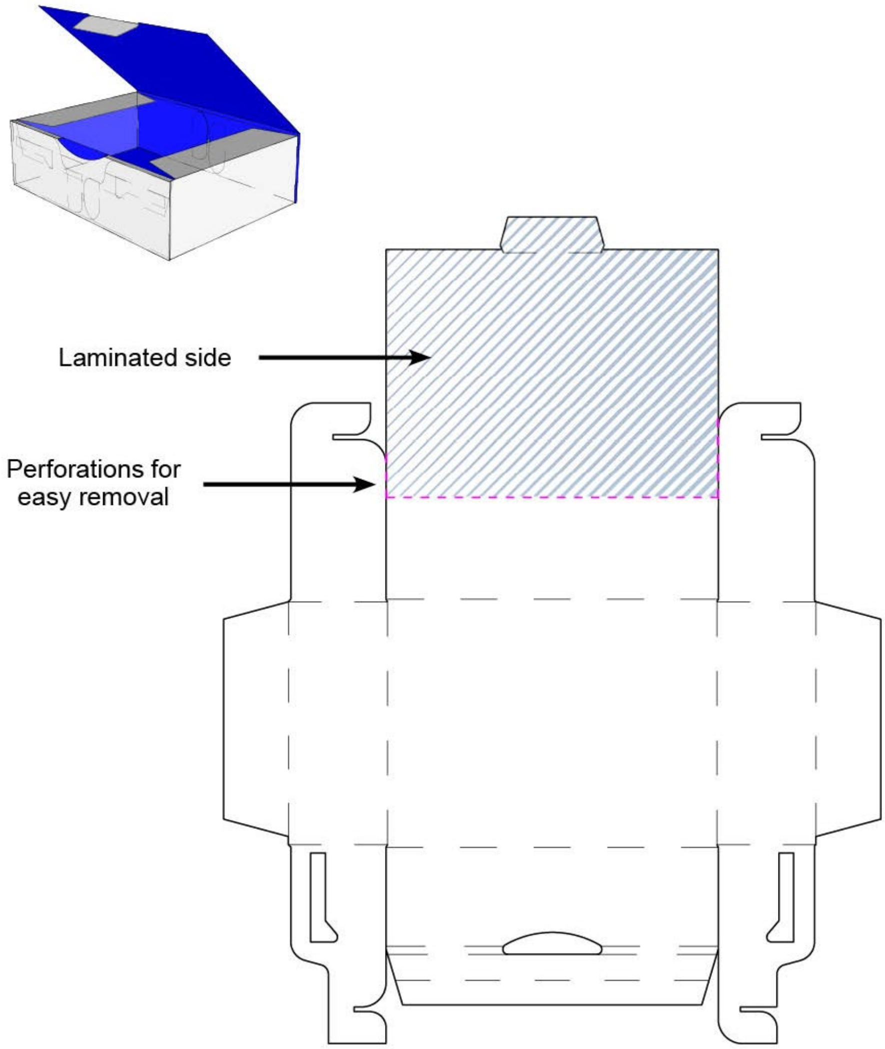

Figure 15. Example of optimized sustainable pharmaceutical packaging

The results of the overall study confirm the importance of a new and optimized design of pharmaceutical packaging. Current design gaps are identified and the need for future research directions is emphasized. The research focus is on complete redesign and its impact on packaging performance. This step is a preliminary design phase where the concept and subsystems are defined using better models to guide design changes and performance expectations are set. The detailed design phase attempts to complete the design down to the smallest detail so that it can eventually be manufactured. All of these phases require iteration within themselves. Therefore, future research will be planned and encouraged based on the guidance gained from this investigation.

11. CONCLUSIONS

The results od the study in Paper III, have confirmed the hypothesis that by examining the production cycle of the pharmaceutical cardboard packaging of the product, it is possible to define points that particularly affect the sustainability of recyceled laboratory paper sheets [264].

- By determining the efficiency of print recycling and characteristics of recycled fibres three methods are compared (P1- without chemicals, P2 - INGEDE 11 and P3 - their combination), concluding that that all methods satisfactorily remove impurity particles from pulp made from printed laminated substrates and printed laminated boxes for drugs, for it is crucial that the paper packaging products contained in the paper for recycling are repulpable within standard operating time and equipment.
- This study has shown that a method without the use of chemical agents can successfully separate impurity particles from paper pulp in a more sustainable manner.
- It is confirmed that specific issue in the separation of impurities is the presence of adhesives used for boxes assembling, as well as silver foil for lamination, showing that there is a higher concentration of adhesive in the pulp when the adhesive is applied to the entire surface of the cardboard or the printed substrate. These substances contribute to the complexity of the process of separating impurity particles from paper pulp because different types of particles are formed (including stickies), many of which can agglomerate due to the presence of adhesive.

The results od the study in Paper I have confirmed the hypothesis claiming laboratory paper and foam handsheets made of paper pulp will contain different concentrations of metal, depending on the manufacturing stages of the recycling process. The data obtained from the research as well as those that are planned to be obtained in subsequent studies can contribute to the selection of materials that do not contribute to the increase of metals in the cellulose pulp. This knowledge can help assess the correct use of recycled cellulose in the production of cardboard packaging in which food or medicines are packaged [265].

- It has been found that the extraction of metals from cellulose pulp is influenced by the factor of using or not using adhesives and the electronegativity of the metal. We believe that the electronegativity factor is related to the process property of deinking flotation, which depends on the hydrophilicity and hydrophobicity of the substance.

- The processes for extracting metals from pulp are significantly influenced by the composition of the adhesive, which should be taken into account in the design of cardboard packaging. Adhesives for lamination have a greater effect on the separation of the mat from the cellulose pulp than the adhesive applied to the edges of the packaging during its assembly.
- In most cases, the deinking flotation method has proven to be a suitable process for the extraction of metals, and the mass fractions of metals measured in the samples do not belong to the categories that would be of concern for human health.

The results od the study in Paper II, have confirmed that the knowledge gained by observing the manufacturing stages of cardboard packaging products combined with studying the quality of the recycled laboratory paper sheets can be used to optimize the design, manufacturing, and recycling of the same product [266]. The circular economy in the production of cardboard packaging products is most evident through the recycling of cardboard products. The sustainable obtaining of raw materials reduces the negative impact on the environment. The procedures determine the recyclability of existing printed material, as well as the possible adhesive and material improvements. An insight into the influence of each stage of production of packaging intended for pharmaceutical products on the properties and characteristics of recycled paper. The results confirm the importance of a new and optimized design of pharmaceutical packaging. Current design gaps are identified and the need for future research directions is emphasized. The research focus is on complete redesign and its impact on packaging performance. The acquired knowledge can be applied in the design phase of a more sustainable product. Designing for recycling will contribute to an increase in the quality of the obtained paper mass, which is directly related to an increase in the productivity of recycling and the sustainability of the packaging production process.

- Before the removal of ink particles by deinking flotation, a greater number of ink particles in laminated samples compared to non-laminated samples is shown, which affected the decrease in measured values of ISO brightness and chromatic coefficient L*. The sources of these impurities are probably agglomerated ink particles that are collected due to the presence of glue in the paper pulp.

- Laminated pulp samples could contain foil fragments, which may increase the values of the optical parameters. However, such foil fragments were successfully separated before making sheets of laboratory paper.
- By studying the stages of packaging production and their recycling, it can be concluded that the composition of the glue significantly affects the formation of agglomerate particles, i.e. sticky particles. Such a process is additionally encouraged by applying glue to the assembly of the packaging. ERIC numbers and image analysis carried out in the research confirm the aforementioned behaviour of ink and glue particles within the paper stock.
- Production can be optimized by packaging design for the production of paper pulp of optimal quality, meaning the production of high-quality recycled cellulose fibres. Taking into account recyclability it is possible to design a more sustainable product. In that manner, the maximum number of recycling cycles can be enabled for paper pulp, which contributes to the circular economy.

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13. LIST OF FIGURES AND TABLES

14. CURRICULUM VITAE

Mia Klemenčić was born 5th of July, 1986. in Zagreb, where she completed primary and secondary school. She graduated from *the Faculty of Architecture, School of Design at the University of Zagreb* in 2012., specializing in Industrial Design. In the same year she enrolled in Postgraduate studies at *the Faculty of Graphic Arts at the University of Zagreb.* In accordance with her preference for preserving nature, she has chosen to focus the research on sustainable design.

In 2015. she started to work at Naklada Ljevak, where she worked as a head of Graphic department, and built a career in Graphic and Editorial design. Throughout the years she has enrolled in a variety of courses to acquire additional skills.

She is teaching at the Rochester Institute of Technology Croatia as an external associate Professor.

List of publications:

I. Klemenčić, M.; Bolanča Mirković, I.; Bolf, N.; Markić, M. Determination of the Mass Fractions of the Heavy Metals in the Recycled Cellulose Pulp. *Polymers* **2024**, 16, 934. doi.org/10.3390/polym16070934

II. Klemenčić, M.; Bolanča Mirković, I.; Bolf, N. The Influence of the Production Stages of Cardboard Pharmaceutical Packaging on the Circular Economy. *Sustainability* **2023**, 15, 16882. doi.org/10.3390/su152416882

III. Klemenčić, M.; Bolanča Mirković, I.; Bolf, N. The efficiency of the separation of impurities from cellulose pulp obtained from pharmaceutical laminated cardboard packaging // *Tehnički vjesnik*: znanstveno-stručni časopis tehničkih fakulteta Sveučilišta u Osijeku, 29 (**2022**), 4; 1295-1300. doi: 10.17559/tv-20210831164929

IV. Bolanča Mirković, I.; Klemenčić, M.; Bolf, N. Karakterizacija čestica na recikliranim listovima papira dobivenim od otisnute kartonske ambalaže za lijekove // Matrib ... (USB) / Šolić, Sanja; Schauperl, Zdravko; Pugar, Daniel (ur.). 2019. str. 52-59

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APPENDICES

List of Publications is summerised in the Thesis.

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Article

Determination of the Mass Fractions of the Heavy Metals in the Recycled Cellulose Pulp

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Special Issue
Advanced Preparation and Application of Cellulose

Edited by
Dr. Marta Fernandes and Dr. Jorge Padrão

MDPI

https://doi.org/10.3390/polym16070934

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Article

Determination of the Mass Fractions of the Heavy Metals in the Recycled Cellulose Pulp

Mia Klemenčić [1,*], Ivana Bolanča Mirković [1], Nenad Bolf [2] and Marinko Markić [2]

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Abstract: In the process of paper recycling, certain amounts of metals can be found in the cellulose suspension, the source of which is mainly printing inks. The paper industry often uses different technologies to reduce heavy metal emissions. The recycling of laminated packaging contributes to the formation of sticky particles, which affects the concentration of heavy metals. This study aimed to determine the mass fraction of metals in the different phases of the deinking process to optimize the cellulose pulp's quality and design healthy correct packaging products. In this research, the deinking flotation of laminated and non-laminated samples was carried out by the Ingede 11 method. As a result of the study, the mass fractions of metals in cellulose pulp were divided into four groups according to the mass fraction's increasing value and the metals' increasing electronegativity. The quantities of metals were analyzed using Inductively Coupled Mass Spectrometry (ICP-MS). The separation of metals from cellulose pulp is influenced by the presence of adhesives and the electronegativity of the metal. The results of the study show that the recycling process removes certain heavy metals very well, which indicates the good recycling potential of pharmaceutical cardboard samples.

Keywords: recycling paper; pharmaceutical packaging; flotation deinking; heavy metals; IPC-MS

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Citation: Klemenčić, M.; Bolanča Mirković, I.; Bolf, N.; Markić, M. Determination of the Mass Fractions of the Heavy Metals in the Recycled Cellulose Pulp. *Polymers* **2024**, *16*, 934. https://doi.org/10.3390/polym16070934

Academic Editors: Marta Fernandes and Jorge Padrão

Received: 1 March 2024
Revised: 21 March 2024
Accepted: 24 March 2024
Published: 29 March 2024

1. Introduction

Cellulose is the most abundant natural polymer on Earth [1]. It is a flexible, renewable, and biodegradable raw material, making it widely used in the packaging industry [2–4]. All plant matter has, on average, a cellulose concentration of roughly 33% [5]. Cellulose is a complex carbohydrate found in plant cell walls, providing rigidity and strength to plant cells. It is made up of repeating units of β-D-glucose linked together by β-1,4-glycosidic bonds. It has a linear semicrystalline structure containing a long chain of repeated D-glucose units linked by a β-1,4 glycosidic bond between D-glucopyranosyl units. The key process of cellulose pulp production is the extraction of cellulose in its fibrous form [6,7]. Most of the paper today is prepared from the cellulose pulp of coniferous trees (spruce and pines), whose three main components are cellulose, hemicellulose, and lignin [8]. Cellulose has fiber-forming properties due to the presence of straight, long, and parallel chains. It provides strength and stability to the paper structure. Hemicelluloses are a group of polysaccharides that can influence characteristics such as paper porosity, absorbency, and printability. Lignin is a complex polymer that is a natural binding component of wood cells that helps hold cellulose chains together [9]. Paper is a flat material made of plant cellulosic fibers, usually mechanical and/or chemical wood pulp, but also recycled fibers, non-fibrous components (minerals and additives), and water [10]. Different paper products will have different compositions [11,12].

Packaging is known as a protective outside layer, which tends to protect its contents from any physical, chemical, or biological hazards. Food and drugs are subject to the same rules and regulations [13]. Cellulosic paper and paper-based materials are one of the oldest and most widely used food contact materials (FCMs) [14,15]; however, they are porous and offer little resistance to the migration of chemical compounds [16,17]. To improve functional barriers against the transfer of various permeates such as moisture, gases, and lipids through the cellulose wall, they are often treated with additives or laminated [18,19]. Therefore, FCMs are made from raw materials and intentionally added substances (IASs) that extend the service life, but also improve production, stability, mechanical properties, and aesthetics [20,21]. Numerous toxic chemicals such as inks, phthalates, surfactants, bleaches, and hydrocarbons are introduced into paper during the production process [10,22]. Raw materials made from recycled paper and cardboard are more likely to contain some heavy metals as well as other chemical additives [23]. Muncke et al. reported that more than 10,000 chemicals are intentionally used in the production of food contact materials [24].

In paper production, pigments are added to improve the structural and surface properties of the paper. Zinc sulfate is used to increase the opacity of special papers, while zinc oxide is sometimes used to make photocopy paper [25]. Zinc and cadmium pigments are additives that give paper fluorescent properties and increase the cohesive strength of paper surfaces. Zinc is also commonly used in papermaking for fine art applications when white pigments are used [26]. Metals such as copper and aluminum are used for engraving on various packaging [27]. Additionally, the quantity of toxic metals can increase when treating corrugated board packaging using dyes that dissolve in water and an acidic platform without previous surface treatment [25]. The main sources of heavy metals are dyes, which mainly consist of conventional inks and pigments, as well as spot inks and Pantone Matching System (PMS) inks [28]. Inks are considered to increase the content of Cd, Pb, Zn, and Cu [28]. It was concluded that most pigments used in printing inks are based on metal compounds of Zn and Cu, including Pb and Cr, which is why they are already banned for food packaging in some countries. Green and yellow packaging contained compounds such as lead chromate, lead sulfate, and lead oxide [29]. Mohammadpour et al. detected harmful metals such as Pb in high concentrations in most pastry packaging made from recycled paper [30]. In another study, different types of packaging and levels of heavy metals such as Al, As, Ba, Cr, Co, Ni, Pb, and V were analyzed, which in some samples exceeded the permitted concentration [31]. Chang et al. developed a new Liquid Chromatography Mass Spectrometry (LC-MS/MS) analytical method. This method was found to be effective in the rapid analysis of photoinitiators with a high degree of reliability [32]. The ICP-MS method developed for determining the mass fractions of chemical elements (Al, Ba, Fe, Mg, Mn, Pb, Sr, Zn) in paper samples proved to be linear and precise. Values of relative measurement uncertainty ranged from 7.7% to 13.6%, and the used approach allows for improving the quality of data and decision making [33].

However, FCMs may also contain unintentionally added substances (NIASs). The recycled paper contains more total NIAS than virgin paper [34,35]. Identification of these substances is often difficult, sometimes impossible [36,37]. For NIAS screening, substances can be divided into three main groups: volatile, semi-volatile, and non-volatile [38,39]. High-resolution precision mass spectrometry (MS) is a valuable tool for the analysis of non-target substances, including screening and chemical identification, and LC-Orbitrap MS is used to screen suspected migrating compounds in paper food packaging [40,41]. Universal detectors that ensure the detection of the largest possible amount of substances are preferred [39,42]. However, there is no single technique for the assessment of trace metals in materials or their migration, and usually several of them must be combined [16]. In packaging that comes into direct contact with food, contaminants can migrate, that is, chemical compounds from the structure of paper and cardboard packaging can move into the food [28]. This requires a comprehensive analysis of all ingredients that can reach toxicological concentrations in food [16].

In recent years, there have been several research efforts to provide recommendations for the safety of chemicals used in specific packaging and to provide a science-based basis for the development of risk management strategies [40,43–47]. For cardboard and paper packaging products, some standards and regulations limit the content of heavy metals and other harmful substances to ensure safety and environmental protection. In the European Union, the Packaging and Packaging Waste Directive (94/62/EC) provides the framework for limiting the presence of heavy metals in packaging [48]. The directive limits the concentrations of lead, cadmium, mercury, and hexavalent chromium in packaging materials to a maximum value of 100 ppm (mg/kg) for the total content of all four metals combined. This standard applies to all packaging within the EU and to imported packaging material. In the United States, packaging regulations are often state-specific, such as California's Proposition 65, which requires labeling of products containing chemicals known to cause cancer, birth defects, or other reproductive harm [49]. Although Proposition 65 does not specifically target packaging, its provisions affect packaging materials used for products sold in California. The circular economy is the primary goal of socially responsible and sustainable businesses, but the aforementioned standards emphasize the importance of knowing the composition of recycled raw materials and their suitability for health.

In this study, the separation of metals from the different stages of deinking flotation, which is carried out using the INGEDE 11 method, from cellulose pulp was investigated. The deinking flotation method is based on the separation of impurities based on their hydrophilicity or hydrophobicity. Cellulose fibers remain in suspension, and hydrophobic impurities and cellulose chains that are shorter in length or are aggregated with adhesives into sticky particles are largely separated in the flotation foam. The study aims to determine the concentration of some heavy metals in a sheet of paper obtained from cellulose pulp and made from cardboard packaging before and after the deinking flotation process. The content of heavy metals in the packaging was analyzed using ICP-MS. The results can be used to determine the quality of the cellulose pulp, i.e., its chemical suitability for use in the production of recycled cellulose. The research results will also show which metals have a lower affinity for separation from the cellulose suspension. If possible, it would be good to omit such metals in the design and production of paper and cardboard products, which can lead to a higher quality of cellulose pulp.

2. Materials and Methods

In this study, printing substrate, printed quire, and printed packaging were analyzed. The printing substrate was GC2 cardboard, to which an acrylic polymer-based dispersion was applied. The cellulose and other components of the cardboard printing substrate meet the conditions for food and pharmaceutical products, which means that it does not contain increased concentrations of metals and other harmful components. This is crucial for analyzing the concentration of metals, which is the subject of this research. The printing substrate is also a sustainable product, as it consists of 60% virgin fibers and 30% high-quality recycled fibers. The front side of the printing substrate is coated with a three-layer pigment varnish, while the back side is only coated once.

The laminated printing surface is produced with a dispersion based on acrylic polymers (biaxially oriented polyethylene terephthalate), also known as metalized BoPET film (Ultralen Film GmbH, Rhein, Germany) on the same GC2 cardboard. These self-crosslinking acrylics are free of plasticizers and alkylphenol ethoxylates (APEOs). The APEO compounds can affect the environment, aquatic organisms, and humans. The data show that short-chain compounds have a much lower impact than long-chain compounds [50]. The function of the dispersion is to apply a biaxially oriented polyethylene terephthalate film (BOPET metalized) to obtain a laminated packaging material. The film complies with the requirements of the repealed Directive 20/590/EEC.

The samples used are laminated and non-laminated substrate, quire, and packaging. The prints were produced on a five-color offset machine with standard UV offset colors from a European manufacturer, white cover offset printing ink, CMYK (cyan, magenta, yellow, black) inks, and dark purple-blue Pantone Matching System (PMS) inks (Zagreb, Croatia). The printing samples were produced using a standard printing form on a Roland 705 five-color offset press machine. The prints were prepared with UV offset inks from recognized European ink factories. The printing form consists of CMYK (cyan, magenta, yellow, and black) and RGB (red, green, and blue) profiles with a raster tonal value of 10 to 100%, full tones of CMYK (cyan, magenta, yellow, and black) and RGB (red, green, and blue colors), and packaging for medicines. The accuracy of the printed form is 83%. The printing process started with white offset printing ink, continued with CMYK separation, and at the end of the printing process, a dark purple-blue Pantone ink was used for the text on the packaging. For rub resistance, the prints were varnished with a UV-curable varnish, a highly reactive photopolymerizable acrylate system that is free of volatile organic compounds (VOCs) and has a low odor and optimal wetting properties. The UV-cured varnish mentioned is VP 1038 high gloss (marked L2). For the assembly of the packaging, an adhesive was applied to the edges that comply with the European Framework Directive 89/109/EEC, the specific regulations for adhesives in the food industry, and the Regulation (EC) of the European Parliament and the Council on materials and articles intended to come into contact with foodstuffs, as well as repealed Directives 80/590/EEC and Commission Regulation (EU) No. 10/2011 on plastic materials and articles intended to come into contact with foodstuffs [51,52].

The samples were disintegrated into cellulose pulp according to ISO 5263-2:2004 [53]. The standard flotation deinking method INGEDE 11 [54] was used to separate the ink particles from the cellulose pulp. The handsheets were produced according to the INGEDE 1 procedure [55] and the ISO 5269-2:2004 standard [56]. The standard handsheets for this study were produced with a device for the automatic production of paper sheets, the Rapid-Köthen-Blattformer sheet former from Frank—PTI, according to the ISO 5269 standard method.

Figure 1 shows Flowchart of the study, exhibiting all steps of the research. In all procedures, sodium hydroxide was added to the cellulose pulp, giving it an alkaline character (pH = 9.0–11.0). This pH value favors the reactions of saponification and/or hydrolysis of resins from printing inks as well as the swelling of cellulose fibers, which makes them more flexible (better separation of the ink particles). H_2O_2 is used to lighten and prevent darkening of the pulp. Therefore, in the procedures with H_2O_2 and Na_2SiO_3, a silicate is included to prevent the decomposition of H_2O_2. The positive properties of Na_2SiO_3 are also evident in the reduction in surface tension, the effect on particle dispersion, and the prevention of the binding of impurities.

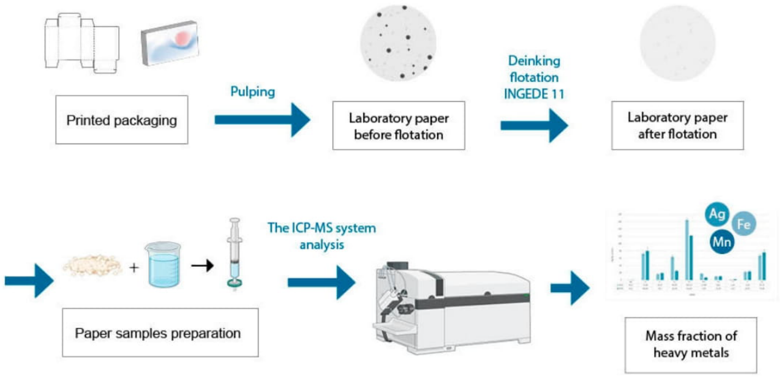


Figure 1. Flowchart of the study.

Table 1 shows samples of laboratory papers made from non-laminated and laminated prints on a cardboard printing base intended for pharmaceutical packaging, before and after the process of separating the ink particles from the cellulose fibers. The quality of the cellulose suspension is also affected by adhesive particles that have formed in the laminated sheet samples and the packaging sample.

Table 1. Samples of laboratory papers made from non-laminated and laminated prints.

Symbol	Sample
P_BF	printed packaging before flotation
P_AF	printed packaging after flotation
Q_BF	printed quire before flotation
Q_AF	printed quire after flotation
S_BF	printing substrate before flotation
S_AF	printing substrate after flotation

The ICP-MS Analysis

The next step of the study was to determine the metal amounts in the laboratory paper and in the cellulose pulp. The method used for the analysis is inductively coupled plasma mass spectrometry (ICP-MS). The ICP-MS system measures the concentration of elements quantitatively and gives the total amount of each element of interest. The process can be divided into four stages: sample feed, ICP torch, interface, and MS.

The recycled paper sample was cut into small pieces and then weighed to approximately 100 mg. To extract the metals from the paper, 5 mL of hydrochloric acid and nitric acid (1:3 ratio) were added to the sample (J.T. Baker, p.a. purity). After dilution, the analysis was carried out; the sample was filtered with a syringe filter and then diluted ten times. Elemental analysis (Ag, Co, Cr, Cu, Fe, Mn, Ni, Ti, V, Zn analysis) was performed by inductively coupled plasma mass spectrophotometry (ICP-MS PerkinElmer SCIEX ™ ELAN® DRC-e, Concord, ON, Canada), which applies continuous scattering (Table 2). ICP-MS is a tool for analyzing trace metals in environmental samples. In this method, the sample is atomized to generate atomic and small polyatomic ions, which are then detected. The working conditions of the device are listed in Table 2. It is possible to detect metals and various non-metals in liquid samples at very low concentrations.

Table 2. The working conditions of ICP-MS PerkinElmer.

Parameters	Working Conditions
Spray gas flow rate	0.85 L/min
Auxiliary gas flow rate	1.2 L/min
Plasma flow rate	14 L/min;
Lens Voltage	8.5 V; ICP RF
Power supply	1100 W; CeO/Ce = 0.016; Ba^{++}/Ba^{+} = 0.015

The ICP-MS calibration was carried out using certified standards. Internal standards are used to compensate for possible measurement deviations. A large number of elements can be detected with an ICP-MS. More than 70 elements can be measured simultaneously in a single analysis. It can measure virtually every naturally occurring element plus many non-natural "radiogenic" isotopes such as technetium, neptunium, plutonium, and americium. The only elements that ICP-MS cannot measure are H and He (which are below the mass range of the mass spectrometer), Ar, N, and O (which are present at high levels from the plasma and air), and F and Ne (which cannot be ionized in an argon plasma) [57]. The advantages of using plasma over other ionization methods, such as flame ionization, are that the ionization takes place in a chemically inert environment, which prevents the formation of oxides, and the ionization is more complete. In addition, the temperature profile of the torch is relatively uniform, which reduces self-absorption effects. Linear calibration curves over several orders of magnitude are observed for ionization processes.

For mass spectrometry, the generation of particles in the submicron range with efficient particle transport to the ICP plays a decisive role [58]. The development and use of the plasma torch as an excitation and ionization source in spectrometry has brought about an important development in analytical elemental analysis. Nowadays, ICP-MS is an essential analytical technique in various fields. This technique requires simple spectra, adequate spectral resolution, and low detection limits for nearly all the elements it can measure. It can detect many elements at levels below 0.1 parts per trillion (ppt). It can also measure elements at concentrations up to 100 s or even 1000 s parts per million (ppm). All metals whose presence was analyzed in this work have a detection limit of up to 10 ppt [57]. For this reason, a mass spectrometer was used as a detector and a high-pressure plasma as an ion source [59]. This method is suitable for the comparison of laboratory sheets, as it is fast, accurate, and precise and allows the analysis of trace elements at low concentrations. The ICP-MS method was used for multi-element analysis for the quantitative determination of silver (Ag), cobalt (Co), copper (Cu), chromium (Cr), nickel (Ni), iron (Fe), manganese (Mn), titanium (Ti), vanadium (V), and zinc (Zn). The mass fraction of all metals is determined according to Formulas (1) and (2).

$$\text{Species mass fraction in the mixture} = Wi = \frac{\text{Mass of specie 'i' in mixture (mg)}}{\text{Total mass of mixture (kg)}} \quad (1)$$

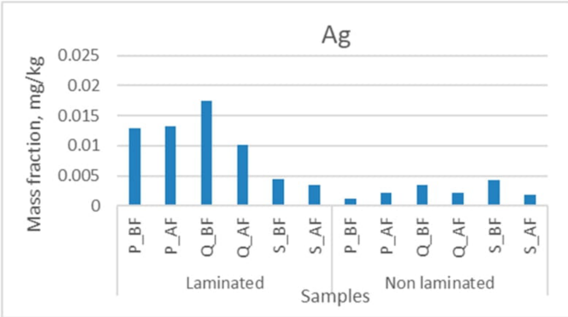
$$\sum_{n=1}^{I=n} Wi = 1 \quad (2)$$

3. Results and Discussion

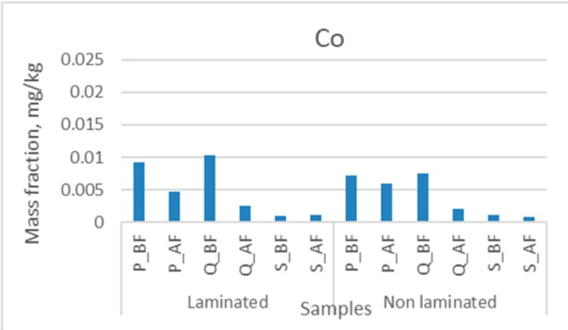
We have divided the measured mass fractions of metals in laminated and non-laminated samples into four groups so that the graphical representation of the results is as easy to read as possible (mass fractions up to 0.025 mg/kg, 0.04 mg/kg, 0.4 mg/kg, and 7 mg/kg). The mass fractions of silver and cobalt in the unlaminated and laminated samples before and after the deinking flotation process are shown in Figure 2. The mass fractions of silver in the laminated samples are higher than those of the cobalt samples, which is due to the lamination of the sheets. Examination of the mass fractions of the metals in the laboratory paper samples from laminated boxes and sheets before and after deinking flotation leads to the conclusion that cobalt is better separated in the flotation foam. The separation of impurity particles by the deinking flotation method depends on many factors. The quality of the paper collected for recycling, the age of the printed product, and the climatic conditions during its life cycle as well as the coatings, varnishes, laminates, and adhesives can affect their deinkability [12,60–65]. Some of these are the small or large size of the ink particles (particles from 10 to 100 μm are best separated), the weak hydrophobicity of the ink particles, the difficulty of separating the particles from the cellulose, the presence of adhesives in the cellulose pulp (formation of sticky particles), and others. The INGEDE 11 method defines the added surfactants sodium hydroxide and sodium silicate, which combine the various necessary functional properties of surfactants and non-surfactants. At the same time, the oleic acid influences the value of the hydrophilic–lipophilic balance (HLB) [66,67]. All this had an impact on the extraction of metal particles from the cellulose pulp, i.e., on the separation from the cellulose fibers. It can be said that the polarity of an atom depends on its electronegativity. In the research, we used the Allred–Rochow electronegativity, which determines the values of the electrostatic force with which the effective nuclear charge acts on the valence electrons. According to the given scale, the electronegativity of the elements is as follows: Ag (1.42), Co (1.70), Cu (1.75), Cr (1.56), Ni (1.75), Fe (1.64), Mn (1.60), Ti (1.32), V (1.45), and Zn (1.66). The value of the effective nuclear charges can be described by the following rule; the higher the charge, the more likely it is to attract electrons. This value is estimated using Slater's rules.

$$Z_{eff} = Z - S \quad (3)$$

Effective nuclear charge (Z_{eff})
Actual nuclear charge (Z)
Shielding constant (S)



(a)

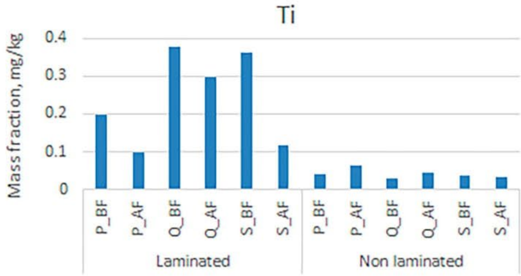


(b)

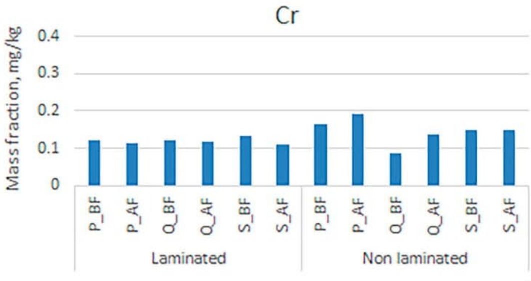
Figure 2. Mass fractions in unlaminated and laminated samples before and after the deinking flotation process for (**a**) silver and (**b**) cobalt.

Cobalt has a higher electronegativity (1.70) than silver (1.42), which could be associated with better extraction methods for cobalt from cellulose suspensions. Figure 2 shows that the mass fraction of cobalt decreases more than that of silver after the deinking flotation process.

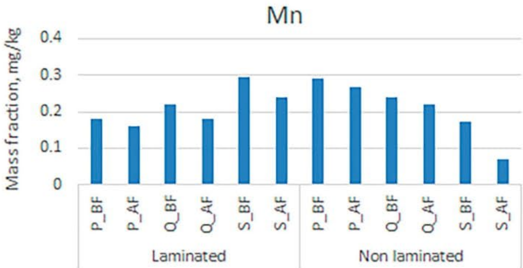
When examining the mass fractions of the metals in Figure 3, it is noticeable that the smallest quantities of nickel were detected. It can also be seen that the concentrations in laminated prints are much higher than in non-laminated packaging. It can be assumed that adhesives significantly influence the presence of the mentioned metals, but nickel can be successfully separated by the deinking flotation process. For the samples of manganese and chromium, there is no significant difference between the mass fraction of the metal in laminated and non-laminated samples, but it must be emphasized that slightly higher mass fractions were detected in non-laminated samples. It can be seen that manganese is better separated during deinking flotation, which could also be attributed to the higher electronegativity (1.60). In this series of metals, the least electronegative metal is titanium (1.32), which was found to have significantly higher mass fractions in laminated samples. Increased concentrations of titanium in laminated samples may be associated with the use of white opaque paints applied to the plastic foil for lamination [68,69]. Sticky particles are formed in the cellulose pulp of the laminated samples, to which titanium can adhere and which also contribute to the increase in the mass concentration values. Confirmation of the above can be seen in the low values of the mass fraction of the non-laminated samples.



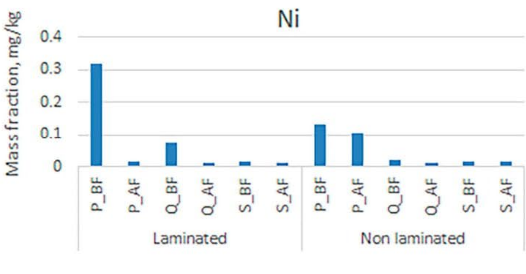
(**a**)



(**b**)



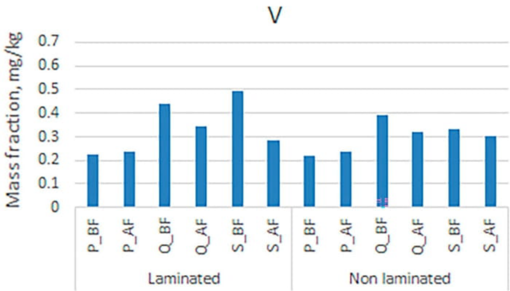
(**c**)



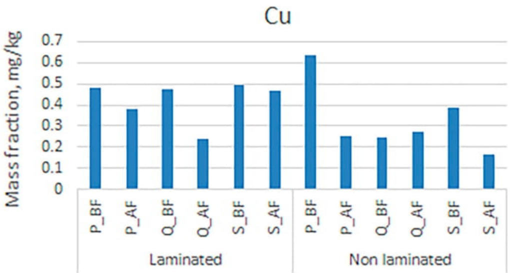
(**d**)

Figure 3. Mass fractions in unlaminated and laminated samples before and after the deinking flotation process for (**a**) titanium, (**b**) chromium, (**c**) manganese, and (**d**) nickel.

Figure 4 shows the mass fractions of vanadium and copper. Slightly higher concentrations of copper were detected in samples of laboratory paper sheets. When examining the mass fractions of copper before and after the deinking flotation process, it can be seen that copper separation is most successful in the sample made from non-laminated boxes. From this, it could be concluded that the adhesive for lamination, which is applied to the entire surface of the printing substrate with a laminated box and quire samples, is more effective than the adhesive for gluing the edges of the box. When examining the influence of the type of adhesive mentioned on the extraction of ink particles from cellulose pulp in the deinking flotation process, the trend is the opposite [70]. When examining the mass fractions before and after the deinking flotation process, it can also be seen that the laminating adhesive has no significant influence on the results. It is likely that the lower electronegativity (1.45) and the adhesive attached to the edges of the box contribute to the formation of larger particles, which contribute to a slight increase in the mass fraction of vanadium after the deinking process.



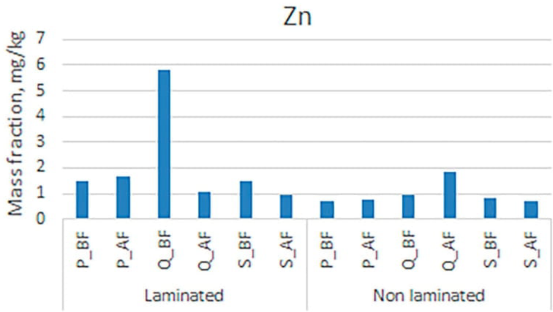
(a)



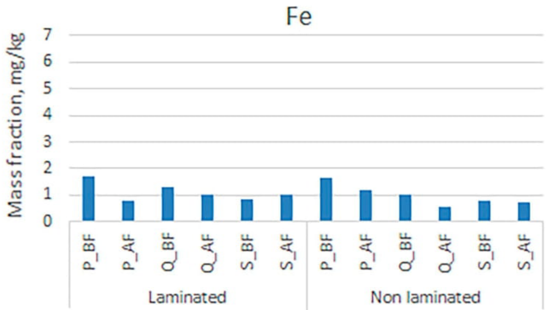
(b)

Figure 4. Mass fractions in unlaminated and laminated samples before and after the deinking flotation process for (a) vanadium and (b) copper.

Figure 5 shows that mass fractions of zinc in the samples are slightly higher than those of iron. It can be noticed that the fat content of zinc increases in non-laminated boxes and quires increase after the deinking flotation process. Since no adhesive is present in the mentioned samples, this behavior of zinc could be explained by the lower electronegativity of zinc (1.66), which may negatively affect the extraction process and contribute to its concentration in the cellulose pulp. The literature does not describe frequent occurrences of negative consequences of elevated zinc concentrations for humans, which can lead to gastrointestinal symptoms, reduced absorption of other minerals (copper and iron), immune disorders, neurological symptoms, liver and kidney damage, etc. The diseases mentioned are more likely to be related to occupational exposure than to contamination of food or packaging [71,72]. The results obtained in this research also confirm the facts from the literature about unlikely effects on human health (the mass fraction is below 3.5 mg/kg). The extraction of iron in the process is not among the most efficient in this study, but it has a constant trend of reducing the mass fraction in the cellulose pulp. It should be noted that in the case of iron, the negative influence of the lamination adhesive on the reduction in the mass fraction could be threatened. In support of this, the results confirm the increase in the mass concentration of iron in the samples after the printing substrate deinking flotation process.



(**a**)



(**b**)

Figure 5. Mass fractions in unlaminated and laminated samples before and after the deinking flotation process for (**a**) zinc and (**b**) iron.

From repeated measurements, silver (Ag), cobalt (Co), copper (Cu), chromium (Cr), nickel (Ni), iron (Fe), manganese (Mn), and titanium (Ti) were calculated for all metals. Vanadium (V) and zinc (Zn) values are presented in terms of standard deviation (SD) and variance (σ^2). Table 3 shows the values for laminated samples, and Table 4 shows the values for non-laminated samples.

Variance is a measure of the dispersion of measured variables, the average sum of the squares of the deviations of the quantity value from the arithmetic mean, while the standard deviation is the positive square root of the variance. Standard deviation is a measure of deviation. It is evident from Tables 3 and 4 that there are no major deviations obtained in the process of determining the mass fractions of metals, which points to the advantage of the chosen method.

Table 3. Standard deviation and variance of the mass fractions of the metals for the laminated samples.

		Ag	Co	Cu	Cr	Ni	Fe	Mn	Ti	V	Zn
Printed packaging before flotation	SD	0.031	0.125	0.025	0.125	0.025	0.045	0.035	0.025	0.055	0.125
	σ^2	0.025	0.101	0.001	0.081	0.001	0.031	0.021	0.001	0.041	0.078
Printed packaging after flotation	SD	0.030	0.106	0.006	0.096	0.016	0.036	0.026	0.016	0.032	0.099
	σ^2	0.021	0.101	0.001	0.071	0.011	0.021	0.021	0.011	0.029	0.077
Printed quire before flotation	SD	0.020	0.085	0.015	0.055	0.025	0.025	0.025	0.025	0.025	0.085
	σ^2	0.015	0.081	0.001	0.041	0.011	0.011	0.011	0.011	0.016	0.074
Printed quire after flotation	SD	0.020	0.076	0.011	0.056	0.016	0.016	0.016	0.016	0.032	0.075
	σ^2	0.015	0.061	0.001	0.041	0.011	0.011	0.011	0.011	0.016	0.068
Printed substrate before flotation	SD	0.025	0.055	0.015	0.035	0.025	0.025	0.025	0.025	0.025	0.042
	σ^2	0.015	0.031	0.001	0.021	0.009	0.011	0.011	0.011	0.011	0.031
Printed substrate after flotation	SD	0.020	0.036	0.006	0.026	0.016	0.016	0.016	0.016	0.022	0.032
	σ^2	0.011	0.021	0.001	0.015	0.011	0.011	0.011	0.011	0.011	0.016

Table 4. Standard deviation and variance of the mass fractions of the metals for the non-laminated samples.

		Ag	Co	Cu	Cr	Ni	Fe	Mn	Ti	V	Zn
Printed packaging before flotation	SD	0.025	0.089	0.025	0.095	0.025	0.035	0.025	0.025	0.045	0.093
	σ^2	0.020	0.077	0.001	0.075	0.001	0.022	0.011	0.001	0.031	0.067
Printed packaging after flotation	SD	0.025	0.086	0.006	0.066	0.016	0.026	0.016	0.006	0.022	0.073
	σ^2	0.011	0.071	0.001	0.041	0.011	0.011	0.011	0.011	0.013	0.056
Printed quire before flotation	SD	0.017	0.065	0.015	0.035	0.025	0.020	0.025	0.020	0.020	0.070
	σ^2	0.011	0.041	0.001	0.021	0.010	0.005	0.001	0.001	0.011	0.055
Printed quire after flotation	SD	0.016	0.056	0.010	0.045	0.015	0.010	0.010	0.010	0.021	0.062
	σ^2	0.010	0.040	0.001	0.032	0.011	0.001	0.005	0.004	0.015	0.044
Printed substrate before flotation	SD	0.020	0.050	0.015	0.030	0.021	0.015	0.020	0.020	0.015	0.035
	σ^2	0.011	0.028	0.001	0.015	0.007	0.007	0.001	0.010	0.005	0.025
Printed substrate after flotation	SD	0.020	0.030	0.004	0.020	0.010	0.011	0.010	0.010	0.015	0.025
	σ^2	0.009	0.017	0.001	0.011	0.007	0.008	0.007	0.008	0.007	0.014

4. Conclusions

This research aimed to investigate the composition of pulp and paper concerning the mass fraction of metals, which can help to assess the correctness of the use of recycled cellulose in the production of cardboard packaging in which food or medicines are packaged. By creating databases, the data obtained from the research as well as those that are planned to be obtained in subsequent studies can contribute to the selection of materials that do not contribute to the increase in metals in the cellulose pulp. The recycling of printed materials such as packaging, newspapers, magazines, or other printed products may contain metals in the composition of the paper. The source of the heavy metals is usually in inks and dyes that are applied to the surface of the printing substrate during the printing process. The possible increase in the concentration of certain metals during multiple cycles of recycling would contribute to the health problems of cellulose pulp for the production of packaging products for food and pharmaceutical purposes.

In the study, it has been found that the extraction of metals from cellulose pulp is influenced by the factor of using or not using adhesives and the electronegativity of the metal. We believe that the electronegativity factor is related to the process properties of deinking flotation, which depends on the hydrophilicity and hydrophobicity of the substance. The study shows that the metals Ag, Ti, Cr, V, and Zn, which have a lower electronegativity, have a smaller increase in mass fraction in some phases after deinking flotation. The influence of adhesives and the formation of sticky particles and the influence on the deinking process were also investigated, as were processes related to the separation of ink particles. The results of this study show that the processes for extracting metals from cellulose pulp are significantly influenced by the composition of the adhesive, which should be taken into account in the design of cardboard packaging. Adhesives for lamination have a greater effect on the separation of the mat from the cellulose pulp than the adhesive applied to the edges of the packaging during its assembly. It must be emphasized that the surface on which the lamination adhesive is applied is much larger, so perhaps the reason for this phenomenon is hidden there. In most cases, the deinking flotation method has proven to be a suitable process for the extraction of metals, and the mass fractions of metals measured in the samples do not belong to the categories that would be of concern for human health.

Author Contributions: All authors contributed to the conception and design of the study. M.K. was responsible for Writing—review and editing, Conceptualization and Visualization, I.B.M. was responsible for Writing—original draft, Data curation and Formal analysis, N.B. was responsible for Methodology and Software, M.M. was responsible for Supervision and Validation. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Data Availability Statement: All data generated or analyzed during this study are included in this manuscript.

Conflicts of Interest: The authors declare no conflicts of interest.

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https://doi.org/10.3390/su152416882

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The Influence of the Production Stages of Cardboard Pharmaceutical Packaging on the Circular Economy

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Abstract: Packaging appearance is important in a competitive market. Designers strive to create products that attract customers and often use laminated packaging, due to the attractive appearance and quality characteristics of the material. The circular economy in the recycling of cardboard packaging helps to reduce waste, saves natural resources and increases the quality of the environment. All of the above contributes to sustainable production, but the quality and properties of the obtained recycled paper materials should not be ignored. Recycling of laminated cardboard packaging often has a negative impact on the quality of recycled paper, due to the formation of sticky particles that can affect the optical properties of recycled paper and the efficiency of the recycling process. This article provides insight into the influence of each stage of production of packaging intended for pharmaceutical products on the properties and characteristics of recycled paper. The standard INGEDE 11 deinking method was used to remove dyes and other impurities from the pulp. The obtained optical results of the characteristics of recycled laboratory sheets obtained from laminated and non-laminated cardboard samples were compared in order to determine the impact of each stage of box production on the quality of the paper pulp. The acquired knowledge can be applied in the design phase of a more sustainable product, and laminated materials can be used in luxury products or to increase the functionality of the packaging. Designing for recycling will contribute to an increase in the quality of the obtained paper mass, which is directly related to an increase in the productivity of recycling and the sustainability of the packaging production process.

Keywords: circular economy; phases packaging production; recycling; optical characteristics

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Citation: Klemenčić, M.; Bolanča Mirković, I.; Bolf, N. The Influence of the Production Stages of Cardboard Pharmaceutical Packaging on the Circular Economy. *Sustainability* **2023**, *15*, 16882. https://doi.org/10.3390/su152416882

Academic Editor: Grigorios L. Kyriakopoulos

Received: 9 November 2023
Revised: 13 December 2023
Accepted: 14 December 2023
Published: 15 December 2023

1. Introduction

The circular economy is based on the economic advantages created by reducing the impact on the environment and reducing resources. Such an approach can be achieved by passing circularity through the 9R Framework' extended classic 3R concept [1]. The 9R Framework' includes recover energy, recycle, reuse, remanufacture, restore, repair, reuse, reduce, rethink and discard. The premise of discarding in a circular economy should be kept to a minimum, but one must not forget to mention the existence of that phase. To reduce the negative impact of the rejection phase, it is possible to combine this phase with the energy recovery phase [2]. The mentioned phase does not contribute primarily to the circular economy (due to the impossibility of uninterrupted energy consumption without major losses), so waste could become a raw material for obtaining energy. The methodology "Circular Economy Product Strategy and Business Model Framework" suggests design and business model strategies to be implemented together [3]. The ReSOLVE framework (regenerate, share, optimize, loop, virtualize, exchange) is a basis for some of the best frameworks as backcasting and eco-design for a circular economy (BECE) [4]. Retrospective planning methodology is useful for complex problems in which current trends are incorporated. Such a planning method can increase the probability of solving ecologically complex issues and predict changes with a strong economic impact [5]. Through a holistic approach, BECE (backcasting and eco-design) introduces the postulates of the circular economy into corporate decision making based on conclusions reached by combining operational and systemic premises [4,6]. The ReSOLVE framework emphasizes technology as the driver of key transformations that will provide new incentives for the adoption of new technologies.

The production of high-quality paper requires a high-tech and specific process. Cellulose is the most important biorenewable, biodegradable and natural biopolymer and an excellent raw material for the development of various sustainable and functional materials, including paper [7,8]. Paper cannot be endlessly recycled because the fibres lose their quality after a certain number of recycling cycles and become unusable [9]. More than 40% of the total paper production is based on the use of secondary fibres [10]. One of the most important prerequisites for quality paper recycling is the paper collection system. The supplied paper must be free of impurities, moisture, etc., in order to produce recycled paper of superior quality [11]. The fractions of waste that are introduced into the paper production process should have as high a proportion of cellulose fibres as possible [12]. There are several methods of collecting paper and packaging waste (PPW); however, the choice of methods can affect the efficiency of recycling and the quality of materials in the further process [13]. Doshi et al. suggested improving communication and collaboration throughout the paper production line, including suppliers, manufacturers and chemists [14]. The end result should be more cost-effective and economical production using inks, coatings and adhesives to facilitate the recycling process. The COLLECTORS project (CP) collected data on separate waste collection systems (WCSs) across Europe [15]. An additional 18 million tons of waste could be collected annually in Europe if best-practice collection strategies are implemented, resulting in a 13% reduction in greenhouse gas production associated with packaging and packaging waste [16].

Extending the lifetime of materials through recycling has become a priority [17,18]. Research by Rahimi et al. [19] aimed to achieve savings by improving the environmental profile of packaging and life cycle assessment (LCA) combined with eco-design. Excessive packaging still leads to excessive consumption of materials and energy, which affects the impact of production and transportation processes [20,21]. Manufacturers and recyclers of paper packaging are committed to using the best environmental practices [22–24]. Recovery efficiency must also be achieved through improvements in sorting and recycling (e.g., replacing mechanical recycling with chemical recycling) [25]. It is also important to reduce the demand for paper and packaging materials (PPMs) by implementing ambitious waste control strategies and prevention campaigns [26].

Paper and cardboard are substrates based on cellulose microfibres that have large surface pores, resulting in a poor barrier effect [8,27]. It is important to ensure the user safety and structural stability of paper-based products by impregnation, lamination or additional top layers of conventional materials (e.g., metal plates or mineral plates). The structure of multi-layer packaging consists of two or more substrates that are joined together with adhesive, usually based on polyvinyl acetate (PVAc) [28]. Biobased coatings and adhesives for paper and cardboard are being developed to improve the barrier potential of these substrates [8,29,30]. In the recycling process of laminated packaging, it is important to choose appropriate adhesives because they can cause problems in the recycling process, which will contribute to the competitiveness of the recycled material [31–33]. Unlike laminated paperboards, mixed office paper waste (MOW) is an excellent source of low-cost, high-quality fibres for the paper industry [34].

In order to avoid problems with the quality of finished paper products, such as stains and holes, non-fibrous components, also known as sticky, should be removed as much as possible in the recycling process. These components may originate from the paper or may be added during the processing or use of the paper to prevent paper breakage [35]. In the paper machine, sticky substances clog fabrics and piles, slowing down the drainage of water from the fibre suspension and, thereby, reducing the efficiency of the process [36]. Various chemical and mechanical methods have been proposed to improve the control of sticky particles, and a new approach has been developed using esterase-like enzymes to break down sticky particles into smaller, less sticky particles [37]. Minimizing the impact of these disturbances on the machine, as well as machine downtime, is one of the most difficult problems in paper production. Putz et al. determined the potential for various applications presented [38]. The importance of adhesive selection is indicated by the fact that there is no definitive way to determine the range of contamination because the variable depends on the sample processing conditions (duration, temperature and applied pressure) [39]. Canellas et al. investigated the migration of adhesive compounds from multilayer layers with the structure of the paper-adhesive film and six potential migrants [40].

In addition to the importance of separating sticky particles from paper pulp, many studies conclude that colour removal is the most important step in wastepaper recycling [41,42]. Currently, paper mills use a chemical process to remove ink from wastepaper, which is generally more efficient and economical in terms of ink removal. The severity of ink removal mainly depends on the type of ink, fibre and printing process [43]. Bolanča and Bolanča concluded that the press model affects the particle size and optical properties of laboratory hand sheets of paper [44]. Runte et al. investigated the recyclability of packaging on a laboratory scale, and a method was developed based on a standard stock preparation system for packaging products. Ink removal by flotation plays a key role in the product quality and cost of the wastepaper recycling process [45].

Designing for recycling involves designing a product that is adapted to a quick, easy and efficient recycling process while minimizing the environmental impact [46,47]. The advantage of this design model over other forms is the possibility of designing a product that adapts to an efficient design process, rather than adapting or designing new efficient recycling processes for specific products. Designers in the aforementioned creative processes use materials that are widely accepted in recycling programs, such as paper, cardboard, glass, aluminium and certain types of plastic [48]. An important design paradigm for recycling is the use of mono-materials or those materials that are easily separated [49,50]. To reduce the impact on the environment, it is best to consider the life cycle of the product, that is, to assess the impact of the material on the environment from extraction to production, use and disposal. The analysis will contribute to the circular economy [51].

This study investigates the influence of drug packaging production stages on the characteristics of recycled laboratory paper. The obtained results can provide guidelines for the model design for recycling. Guidelines in the product design phase help design products that will be recycled more efficiently. The mentioned progress enables the use of usual recycling technologies, which would reduce financial expenses for production due to the introduction of new technologies, but, at the same time, production would become more sustainable with higher-quality raw materials produced. This paper will examine how certain stages of production affect the optical characteristics of sheets of recycled laboratory paper, that is, how the use of non-laminated, laminated, printed, non-printed, glued and non-glued cardboard products affects it. The circular economy in the field of cardboard recycling can be implemented in several ways, the basic one being recycling, i.e., establishing a closed circle of materials. Recycling of packaging products can contribute to the circular economy because, from 1991 to 2020, the consumption of paper and cardboard for packaging products grew from 42.1% to 60.1% [24]. For cardboard packaging, manufacturers and distributors can work with recycling companies to ensure a continuous supply of cardboard material and contribute to the sustainability of cardboard production [52]. The process of recycling cardboard materials is not possible indefinitely due to the shortening of the cellulose fibre in each recycling process until it can no longer be used. Usually, recycling is possible up to seven times. An additional reason for the reduced usability of the cardboard material is the presence of glue in the pulp, the source of which occur during the production of laminated material and the application of glue for binding the packaging, which contributes to the formation of sticky particles. By researching the impact of each stage of cardboard packaging production on the quality of the paper pulp, i.e., the recycled sheet of paper, we aimed to apply another circular economy method, design for recycling. The design for the recycling method was used with the premise of designing a product that would yield the highest-quality pulp possible so that it could be used in as many cycles as possible. All methods used in this research are based on the idea that materials and products should move through the system of economic value without creating unnecessary waste.

2. Materials and Methods

Unlaminated and laminated printed packaging, printed quire and substrates were used in this research (Table 1). The same basic printing substrate GC2 cardboard was used for all samples. For the laminated samples, a biaxially oriented polyethylene terephthalate film (BoPET metallized) with plasticizer-free acrylic polymer adhesive or APEO was applied to the basic printing substrate. The APEO compounds can affect the environment, aquatic organisms and humans. The data show that short-chain compounds have a much smaller impact than long-chain compounds [53].

Table 1. Samples used for recycling and their labels.

Type of Sample	Label	Sample	Label
Laminated	L	Printed packaging	P
		Printed quire	Q
		Printing substrate	S
Non-laminated	N	Printed packaging	P
		Printed quire	Q
		Printing substrate	S

The standard prints were produced on a five-colour offset machine: Roland 705 with standard UV offset colours from a European manufacturer, white cover offset printing ink, CMYK (cyan, magenta, yellow, black) inks and dark purple-blue Pantone ink. For rub resistance, VP 1038 high-gloss (product code), UV-cured VergamGH varnish (labelled L2) was used, a highly reactive, photopolymerisable, VOC-free acrylic system with reduced odour and optimal wetting properties. The adhesive used for the assembly of the pharmaceutical packaging was suitable for food packaging and food contact materials. The above materials and equipment were used to prepare samples according to the described packaging stages for further experimentation (Figure 1).

The samples were disintegrated into cellulose pulp according to ISO 5263-2:2004 [54] standard. The standard flotation deinking method INGEDE 11 [55] was used for the separation of the ink particles from the cellulose pulp. The prints are converted in alkaline conditions to pepper pulp, which is subjected to a single flotation process to remove the ink particles. Usually, the sizes of the resulting fragments of printing inks are suitable for separation from cellulose fibres based on their hydrophobicity and hydrophilicity. The handsheets were produced according to the INGEDE 1 procedure [56] and the standard ISO 5269-2:2004 [57]. The standard handsheets for this study were produced using a Rapid-Köthen sheet former. Some of the optical properties of the laboratory handsheets were measured according to the standards listed in Table 2.

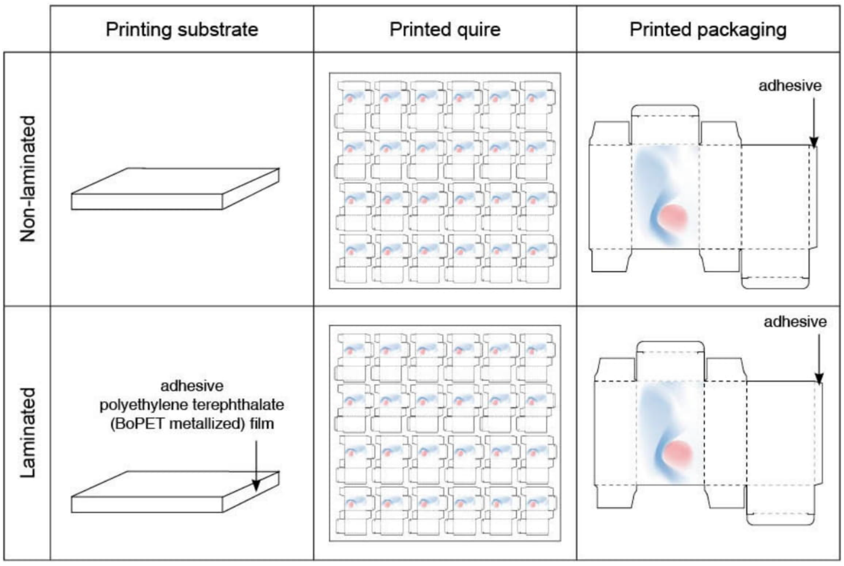

Figure 1. Packaging preparation stage in the experiment for non-laminated and laminated samples.

Table 2. Optical measurements performed on the laboratory handsheets and the standard methods employed.

Optical Properties	Standard
Diffuse blue reflectance factor	ISO 2470-1:2016 [58]
Effective residual ink concentration, ERIC	TAPPI T 567: 2009 [59], ISO 22754:2008 [60]
Determination of colour by diffuse reflectance	ISO 5631-3: 2015 [61]
Image analysis	ISO 13322: 2014 [62]

Spec*Scan Apogee System image analysis software (v2000) was used to determine the count and surface area of the remaining dirt particles. A scanner digitised the images with the following settings: Threshold (100), White Level (75) and Black Level (65) [62].

The spectrophotometer Technidyne Color Touch 2 was used to analyse the effective residual ink concentration (ERIC number) and CIE L*, a*, b* chromatic coefficients on laboratory handsheets, before and after deinking flotation [59–61]. The effective residual ink concentration on samples is measured according to standard methods ISO 22754:2008 and TAPPI T 567: 2009 [59,60]. The standards according to which the chromatic coefficients were measured are shown in Table 2.

3. Results and Discussion

Brightness is defined as the diffuse reflectance of a thick stack of paper when visible light has a wavelength of about 457 nm and a bandwidth of about 40 nm. Such an average wavelength of visible light can be defined as blue light; the human eye perceives a bluish shade as whiter than neutral white in the colour spectrum. The highest brightness is 100, higher brightness is associated with brighter papers and brighter papers are considered premium papers. The brightness parameter does not describe the measurements of other wavelengths of light; samples of different colours can have identical brightness. That is why the colour coefficients were examined in the continuation of the research. The brightness of the paper can affect print quality.

Paper whiteness measures the reflectance of all wavelengths, making it more in line with how the human eye perceives paper. The measurement parameter has a subjectively perceived property, and most people consider it to increase when the material has a slightly blue tint (ISO 11475:2017) [63]. The highest print quality is determined by high whiteness, especially ISO with whiteness from 140 to 175 [64]. Yellow hue is defined as a measure of the degree of change in surface colour from the preferred white (or colourless) to yellow. White titanium dioxide ink is applied to recycled papers to achieve optical properties equal to those of commercial papers recommended in ISO 12647 [65,66].

The measurement under the described conditions is shown in Figure 2. The values determined for the ISO for the brightness of the laboratory paper sheets were lower for the laminated samples. The paper pulp of laminated samples contains a larger number of ink particles and dirt, which negatively affects the brightness of the paper. The described trends of higher ISO brightness values are 0.5% before and 3.5% after flotation for packaging and 0.5% before and 0.3% after flotation for printing substrate. Although lower brightness values were obtained for the laminated samples of laboratory sheets, the values obtained are satisfactory for reuse by making new packaging products. It should be emphasized that in this research, cardboard was recycled, which has lower brightness values than office paper or paper intended for printing representative books. There is a deviation from the described trend in the research results for the printed quire: the brightness of the unlaminated sample increases compared to the laminated sample but it is still higher than the unprinted printing substrate. In the described example, a decrease in the brightness value of 1.5% before and 0.5% after flotation can be observed. In addition to the aforementioned ink and dirt particles that affect the brightness of the paper in the measurement area, the measured values are affected by some other factors. In the mentioned area, the obtained paper reflectance values can be based on chromophores that are usually found in cellulose fibres such as lignin and its by-products [67]. These organic compounds usually absorb the strongest blue wavelengths, resulting in a yellowish colour to the cellulose pulp. The colour of the pulp depends on the life cycle of the pulp, i.e., bleaching, exposure to light, exposure time or ageing, raw material, fibre formation, etc. It can be assumed that some of the above reasons affected the brightness of the sample, especially because the printing pad is made of fifteen layers of paper (Figure 3). It should also be emphasized that these are small value increases of 1.5% before and 0.5% after deinking flotation.

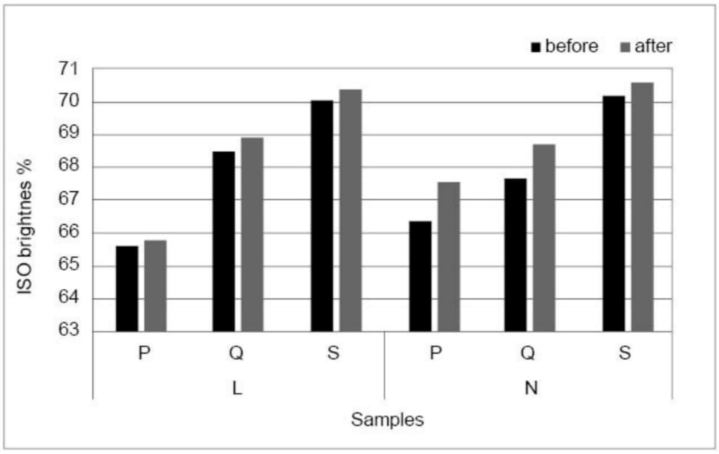

Figure 2. The influence of flotation deinking of laminated and non-laminated samples on ISO brightness.

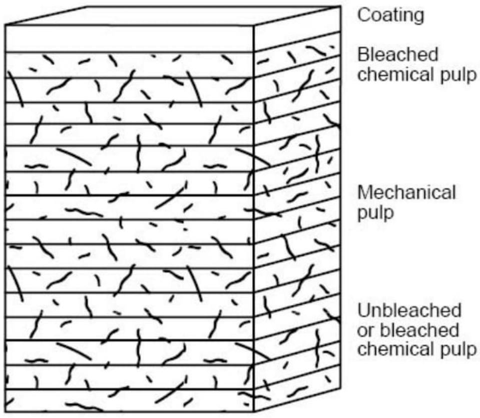

Figure 3. Multiply construction for Folding Packaging Board (FBB) GC2 cardboard.

CMYK (cyan, magenta, yellow, black) colour models were used in the production of the samples. To obtain absolute data that are independent of the measuring device, the CIELAB model/system is used. The CIELAB colour characterization system is used in graphic technology and art. In the CIELAB colour characterization system, three basic dimensions of the perceptual colour attributes, hue, saturation/chroma and lightness, or the three dimensions also known as tristimulus data, are defined [68].

The lightness of a colour laboratory sheet describes its relative brightness, apropos its luminous intensity. The lightness value of samples indicates how light or dark a sheet of paper is, and this indicator is achromatic [69]. One of the parameters affecting the lightness is the printing substrate.

Lightness L* was highest for the substrate samples, both laminated and non-laminated cardboard, which is to be expected as there were no ink particles on the samples. The values obtained are related to the cream-coloured back of the printing substrate and the layers from which the aforementioned substrate is made Figure 4.

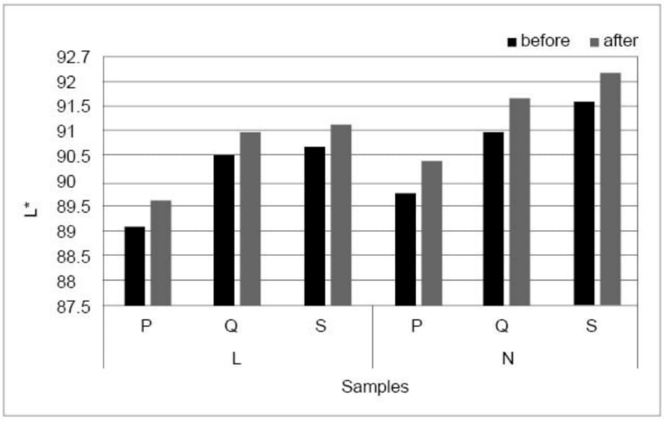

Figure 4. The influence of flotation deinking of laminated and non-laminated samples on the lightness.

The study of the influence of printing on the quire of the sample shows that the laminated samples have a lower difference in the absolute value of the brightness, but the results for the brightness of the substrate of the samples are lower due to the agglomeration caused by the adhesive application. When considering this stage of packaging product manufacture, it should be noted that the adhesives act in the first stage of sample manufacture, and the ink particles only further contribute to the agglomeration effect.

The greatest changes in brightness values occur at the stage when the paper laboratory sheets are produced from the packaging, indicating the effect of adhesive on the packaging ends. In laminated packaging, the larger surface area is coated with the adhesive due to the applied foil than the surface at the edge due to the assembly of the packaging, but Figure 4 shows that the assembly has a greater influence. It can be concluded that the composition of the applied adhesive is extremely important for the properties of recycled paper sheets and their optical characteristics. The resulting sticky particles, together with the ink particles already present in the pulp, form accumulations that contribute to the reduction in brightness for both laminated and non-laminated samples [36,70]. The difference in brightness for the non-laminated samples at this stage is 2.5-times less than for the quire samples, which only confirms the importance of the impact composition of the adhesive applied to the edge of the packaging.

A colour space system is a sequential or continuous representation in which the colour coefficients a* and b* are represented as a* green/red colour component and b* blue/yellow colour component (Figure 5a) [71]. The obtained measured values of chromatic coefficients a* and b* show that they are in the yellow-green area, where the green component is not significantly expressed. Considering the producer's description of the printing substrate mentioned in the methodical part of the paper, it can be concluded that the colour of the printing substrate significantly affects the colour of laboratory sheets of paper that have a yellow tint.

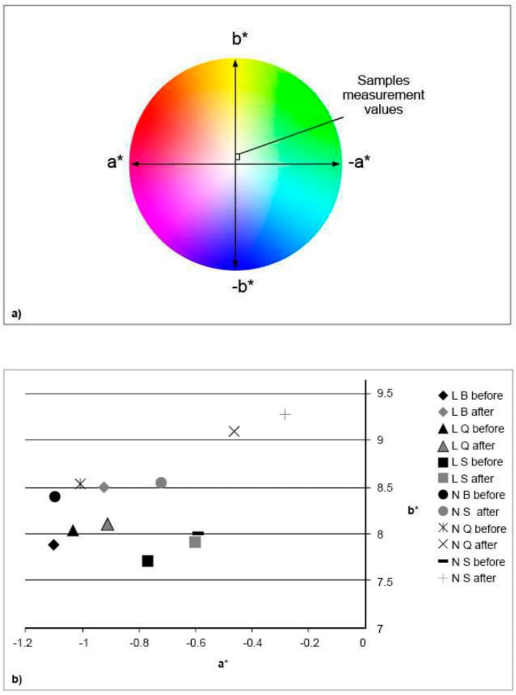

Figure 5. (**a**) Display of the measured values on the CIE ab graph. (**b**) The influence of the flotation deinking of laminated and non-laminated samples on the chromatic coefficients a* and b*.

Since paper pulp originated from packaging products and can be used again to make the same products, it was not necessary to add additional bleaches to the pulp after the deinking-flotation process. The paper pulp can also be used for packaging pharmaceutical products, so bleaching is not even desirable. Raw materials for manufacturing packaging for pharmaceutical products should be treated with as small an amount as possible of chemicals so that they do not affect the product inside the packaging (migration processes). It is considered that the pulp is of higher quality if it is not bleached with chlorine and its compounds. The use of molecular chlorine for bleaching results in the formation of chlorinated organic compounds that are a risk to the environment and the food chain. Chlorine dioxide releases smaller amounts of organochlorine [72]. In recent times, bleaching has been done bycarried out using oxidizing agents, such as oxygen, ozone, sodium hypochlorite and hydrogen peroxide. As this research studies the raw materials for making packaging that do not need and usually are not of great whiteness, there is no need to apply bleaching procedures.

When examining the properties of the sheets after the deinking-flotation process of the prints produced by offset printing with CMYK inks and dark purple-blue Pantone ink, it is obvious that the dye particles were separated from the paper pulp. The laboratory sheets do not contain a large amount of dominant dark-purple-blue dirt particles that would give the sheets the colour mentioned. The description of the green hue on the sheets corresponds to the residual ink particles, over which there are cellulose fibres. This results in yellow-green colouring. The mentioned hue is more pronounced on the samples before the deinking flotation process when the ink particles have not yet been separated from the paper pulp from which the laboratory paper sheets are made (Figure 5a). Therefore, the relationship between the colour and the sign of coefficients a* and b*, or −a* degree of greenness and +b* degree of yellowness, is easy to read [71].

Figure 5b shows that the values of the chromatic coefficient a* are lower for the laminated samples. These results explain that particles of blue-purple printing ink remaining on the sheet before and after flotation impart such a tone to the handsheet, as the separation of the dye particles from these handsheets is less efficient. The substrate samples have a higher coefficient a*, while the laminated samples were yellower and became even yellower after the flotation-deinking process. All non-laminated samples also showed a stronger yellow colouration after the flotation-deinking process.

Laboratory paper sheets made from the laminated sample before and after the deinking-flotation process have higher values of effective residual ink concentration (ERIC) compared to the values of non-laminated samples. Residual ink particles are more efficiently removed from unlaminated paper pulp samples (packaging 22%, sheet 52%) than from pulp made from laminated samples (packaging 16.7, sheet 18.9%). From the results, it can be concluded that the presence of adhesives in the pulp has a significant effect on reducing the extraction of ink particles from the pulp. The greater amount of adhesive in the pulp originates from the lamination of the cardboard material; however, the stage of applying the adhesive to the packaging additionally affects the reduced extraction of ink particles. Earlier measurements of this lightness research in Figure 4 also indicated the importance of the composition of the adhesive on the separation of titanium particles, that is, on the formation of sticky particles. By studying both optical parameters, it can be concluded that the application of adhesive on the edge of the packaging has a greater effect on ink separation (Figure 6) [73].

Image analysis of the handsheet surfaces of the laminated samples before the flotation-deinking process showed a higher number and larger surface area of dirt particles in all classes compared to the non-laminated classes. Figure 7 shows that the higher number and larger surface area of particles were particularly evident in the process before flotation, as already noted in the analysis of the optical properties of the handsheets. It is assumed that stickies with a larger surface area are formed, which are more difficult to separate by flotation deinking, as can be seen in Figure 8 [74]. Understandably, the unprinted substrate samples had a lower number of particles, but it should be noted that large particles still formed in the laminated samples due to the adhesive on the board. The deinking process is more efficient in the non-laminated samples than in the laminated samples, which shows up in the image analysis as a significant difference in the number and surface area of particles on the handsheets before and after flotation deinking.

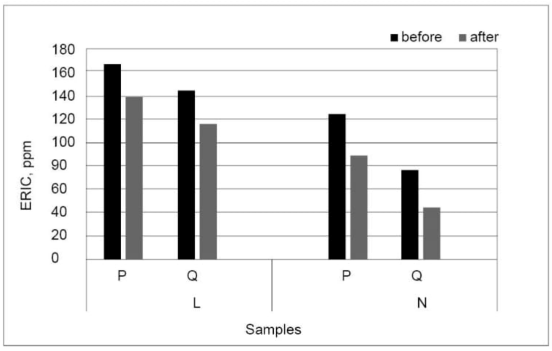

Figure 6. The influence of flotation deinking of laminated and non-laminated samples on ERIC numbers, ppm.

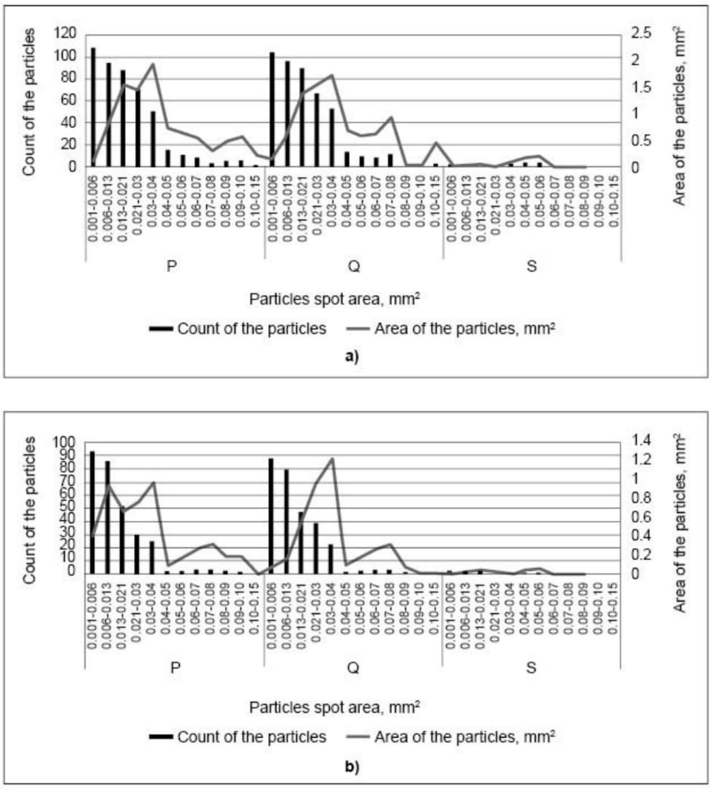

Figure 7. Count and area of particles on the handsheets formed from the laminated samples: (**a**) before deinking flotation, (**b**) after deinking flotation.

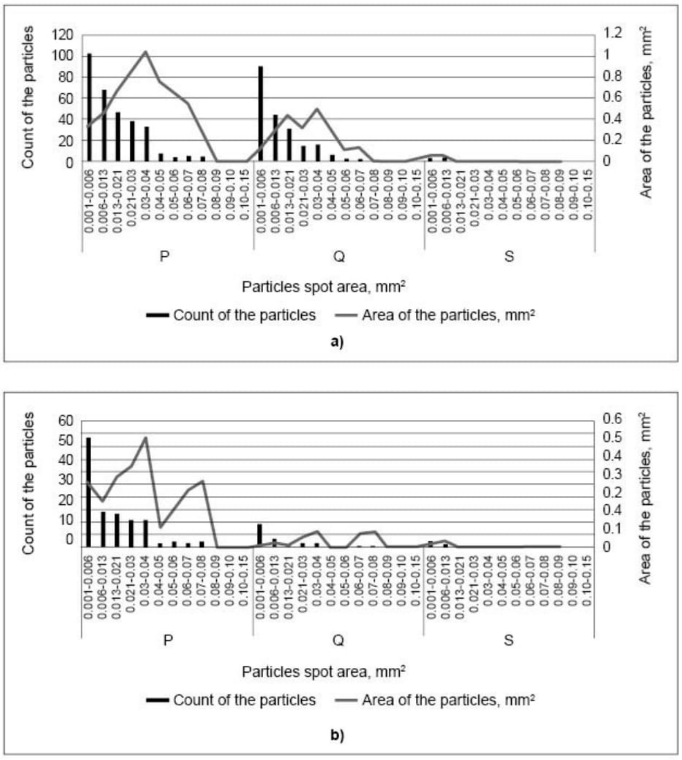

Figure 8. Count and area of particles on the handsheets formed from the non-laminated samples: (**a**) before deinking flotation, (**b**) after deinking flotation.

4. Conclusions

The circular economy in the production of cardboard packaging products is most evident through the recycling of cardboard products. Sustainably obtaining raw materials reduces the negative impact on the environment. Designers of cardboard packaging products in the product design phase can additionally contribute to increasing sustainability with the Design for Recycling (DfR) method [75,76]. The mentioned method is a way of approaching engineering and product design through design, which aims to facilitate the recycling process and increase the utilization of materials in the product. By studying the influence of individual stages of the production of pharmaceutical packaging on the optical properties of recycled paper sheets, insights are gained that can be applied in the product design phase. Pre-consumer post-press waste materials come into the recycling process before glueing the packaging, giving quality raw materials. Post-consumer packaging cardboard has an adhesive applied for glueing the package, which affects the quality of the paper pulp, that is, the formation of sticky particles in the pulp. Laminated materials can contribute to many functional and visual aspects of packaging, but they can reduce the quality of the raw material. It would be good for designers who respect the DrR premise if they have knowledge about the influence of the stages of the lamination process, printing and finishing processes on recyclability.

The research results showed, before the removal of ink particles by deinking flotation, a greater number of ink particles in laminated samples compared to non-laminated samples. The described trend affected the decrease in measured values of ISO brightness and chromatic coefficient L*. The sources of these impurities are probably agglomerated ink particles that are collected due to the presence of glue in the paper pulp that creates sticky particles. Laminated pulp samples could contain foil fragments, which could increase the values of the optical parameters. However, such foil fragments were successfully separated before making sheets of laboratory paper.

Lower efficiency in separating dirt particles in laminated samples is due to the presence of glue in the paper suspension, which leads to particle agglomeration. ERIC numbers and image analysis carried out in this research confirm the aforementioned behaviour of ink and glue particles within the paper stock. By studying the stages of packaging production and their recycling, it can be concluded that the composition of the glue significantly affects the formation of agglomerate particles, i.e., sticky particles. Such a process is additionally encouraged by applying glue to the assembly of the packaging.

Considering all aspects studied, production can be optimized by packaging design for the production of paper pulp of optimal quality, that is, by the production of high-quality recycled cellulose fibres. It would be good if, in addition to other properties of the material, recyclability is mentioned so that designers can come up with a more sustainable product. In the mentioned way, the maximum number of recycling cycles can be enabled for paper pulp, that is, it can contribute to the circular economy. Our further research plans include investigating the opacity and L*a*b* values with a fluorescent filter to gain insight into the loss of cellulose fibres and optical brighteners.

Author Contributions: Conceptualization, M.K. and I.B.M.; methodology, I.B.M.; validation, I.B.M.; formal analysis, M.K.; investigation, M.K.; data curation, M.K. and N.B.; writing—original draft preparation, M.K. and I.B.M.; writing—review and editing, M.K. and I.B.M.; visualization, M.K. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data are contained within the article.

Conflicts of Interest: Author Mia Klemenčić are employed by Naklada Ljevak d.o.o. Other authors declare no conflict of interest.

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ISSN 1330-3651 (Print), ISSN 1848-6339 (Online)
https://doi.org/10.17559/TV-20210831164929
Original scientific paper

The Efficiency of the Separation of Impurities from Cellulose Pulp Obtained from Pharmaceutical Laminated Cardboard Packaging

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Abstract: On the market there are increased quantities of laminated cardboard packaging with glossy surface. Laminated packaging is being used for its appealing appearance. However, laminated paper treatment creates additional problems in the process of recycling of used product. Adhesives and foil are a specific issue in the separation of impurities in recycled paper. It is crucial that the paper packaging products contained in the paper for recycling are repulpable within standard operating time and equipment. In this research three methods (without chemicals, INGEDE 11 and their combination) of removing impurities from paper pulp are compared and their efficiencies are determined to gain deeper insight into the recycling process. The procedures determine the recyclability of existing printed material, as well as the possible adhesive and material improvements. A specific issue in the separation of impurities was the presence of adhesives as well as silver foil (BOPET metallized film) for lamination. All three applied procedures successfully separate impurity particles from paper pulp and printed laminated boxes in a more sustainable manner, without the use of chemical agents.

Keywords: adhesives; deinking; flotation; *L* a* b**; paper and cardboard; pharmaceutical packaging; pulp; recyclability; stickies

1 INTRODUCTION

The evolution of the pharmaceutical industry has had an impact on people's lives because it has contributed, and still contributes to generating well-being in terms of health and quality of life for individuals, and because it is one of the most relevant sectors for the economy at a global level [1]. The pharmaceutical industry has been the subject of growing attention for the impact in terms of sustainability of its activities. Concerns about the introduction of sustainability practices into waste recycling, the reduction of water usage, greener manufacturing methods, and recyclable packaging have intensified attention on this topic. The sustainability of the pharmaceutical industry has also aroused the interest of scholars from various disciplines, such as chemistry, engineering, and environmental sciences [2].

Paper recovery rates continue to increase each year. The American Forest & Paper Association has launched its Better Practices Better Planet 2020 initiative, establishing an ambitious goal of 70% paper recovery by 2020 (the recovery rate was 63.5% in 2010) [3]. However, the use of the recycled paper is still limited by the presence of many kinds of contaminants, which can be classified according to their source, i.e. organic, inorganic and microbiological ones [4].

It is crucial that the paper packaging products contained in the paper for recycling are repulpable within standard operating time and equipment; otherwise too much material is lost and cannot be integrated in the recycled pulp. It is important that the resulting recycled pulp is optically and mechanically homogeneous, which ensures recycled pulp for high quality products. Also, it is important that adhesive impurities do not lead to microstickies at all nor to a macrosticky area that is too big. The most important parameters are therefore repulpability, yield of fibrous material, coarse reject, flake content, stickies and technical quality [5].

Paper sheets are used widely for many purposes. The production over the entire world depends mainly on forest tree pulps, which adversely affects the environment [6]. During traditional pulping processes many hazardous chemicals which damage the environment are used to produce pulp. The technological process of waste paper deinking is composed of four basic units: (1) defibering (preparation of used paper suspension), (2) washing or flotation (removal of impurities from the suspension), (3) bleaching of fibers and (4) treatment of the process water which is used in the recycling of the paper [7, 8]. Deinking is the most important step in waste paper recycling [9-11].

On the market there are increased quantities of laminated cardboard packaging with glossy surface. Laminated cardboard packaging creates additional problems in the recycling process of the used product. The number of sticky particles, but also the shiny particles is greatly increased. In our research typical papers are characterized and compared the particles of impurities on the surface of the handsheets made from recycled fibres obtained from cardboard and laminated cardboard packaging [12].

The heterogeneous nature of the produced wastes depends on the type of recycled paper and on the kinds of process units from which they were obtained. Accepted fractions should be as rich as possible in cellulose fiber up to the paper machine, thus becoming incorporated in the papermaking process [13]. Secondary fibers are the principal feedstock for paper production worldwide due to environmental and economic issues, regarding the use of recycled materials rather than pure cellulose. It was reported that 40% of total paper production is based on the use of such fibers [14].

Various methods are available for the quantification of stickies and can be classified into methods that measure the quantity, composition, or deposition tendency [15]. There are also morphological determinations using screening and microscopic analysis, which can be helpful for macrostickies. In the case of colloidal and microstickies, chemical analysis based on solvent extraction procedures and gravimetric analysis can be used. There are also reports on thermogravimetric analysis (TGA), Fourier-transform infrared spectrometry (FTIR), and nuclear magnetic resonance spectrometry (NMR) studies [16]. When the water fraction is evaluated, it is possible to use a fluorescent counting method [15], or turbidimetry.

However, there is no reliable and repeatable method that is accepted as standard.

The heterogeneous nature of organic contaminants trapped in recycled paper requires an integrated chemical approach for their complete characterization [18] and for the evaluation of their subsequent alternative uses. Some of the most commonly adopted methods of analysis are reported in the literature [19, 20]. Up to now, these methods have been mainly focused on the macrostickies analysis, but there is no generally accepted standardized method for microstickies determination.

The present paper compares three methods for paper recycling by defining recyclability of laminated paper containing adhesives, to determine the efficiency of the methods and to give an insight to the optimization of the recycling process. The research contributes to better understanding the characteristics of the obtained secondary fibers, as well as the classes of impurities which are mainly contained in the cardboard pulp obtained from laminated cardboard for packaging of pharmaceutical products. The aim of the author is to conduct research to determine which process is most effective for separating impurities from the cellulose pulp of laminated cardboard while reducing the negative effects on the environment.

2 EXPERIMENTAL

In this study, laminated cardboard printing substrate, labelled here as S, and printed pharmaceutical laminated cardboard packaging, labelled here as P, are used. Printed substrate was the standard sample for determining the efficiency of the deinking process. The printing substrate was GC2 cardboard, to which a dispersion based on acrylic polymers was applied. These self-crosslinking acrylics are free of plasticizers and alkylphenol ethoxylates (APEO). The function of the dispersion is to apply a biaxially-oriented polyethylene terephthalate (BOPET metallized) film to obtain a laminated packaging material. The BOPET metallized film complies with the repealed Directive 20/590/EEC. Printing samples were made with a standard printing form on a five-color offset machine, Roland 705. The prints were prepared with UV offset inks produced by Sun Chemical® Europe. The printing process started with white offset printing ink, continued with CMYK separation and a dark purple-blue pantone color was used at the end of the printing process for the text on the packaging. The prints were varnished with a UV-cured varnish, which is a highly reactive photopolymerizable acrylate system, VOC free, with reduced odor and optimal wetting properties. The aforementioned UV-cured varnish is VP 1038 high gloss, VergamGH (marked L2). For assembling the packaging, an adhesive was applied to the edges in compliance with the European framework directive 89/109/EEC, specific rules for adhesives in food applications and regulation (EC) of the European Parliament and of the Council on materials and articles intended to come into contact with food and repealing Directives 80/590/EEC, and Commission Regulation (EU) No 10/2011 on plastic materials and articles intended to come into contact with food.

After conversion of the sample into cellulose pulp (decomposition process) according to ISO 5263-2: 2004 [21], separation of impurity particles was performed in three ways.

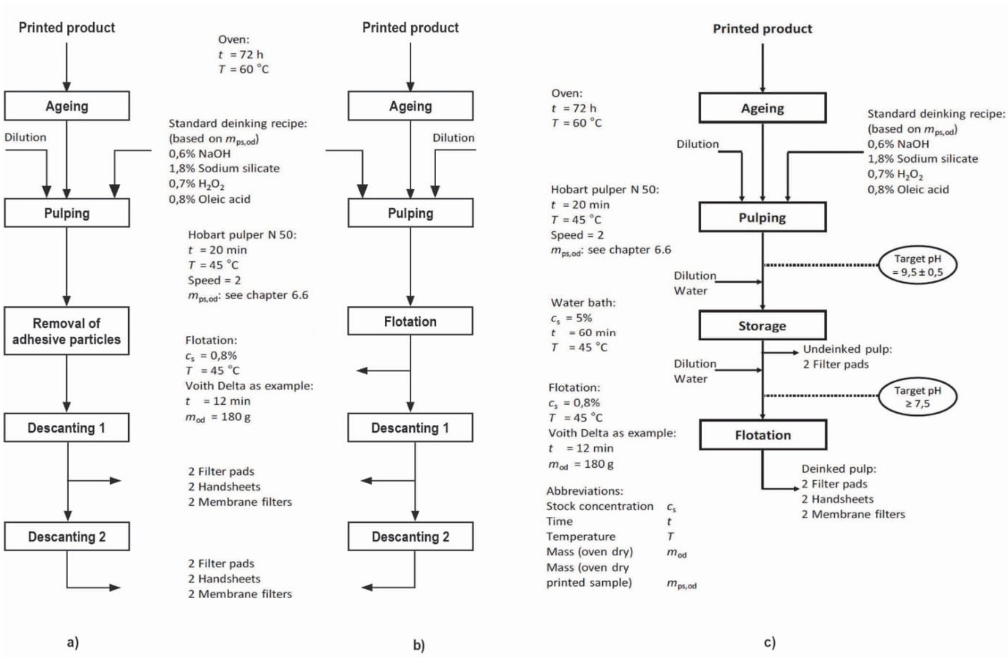

Figure 1 The flow diagram of a) a method without the utilization chemicals b) a combined method c) and the INGEDE Method 11 [22]

The standard INGEDE method 11 of deinking flotation was used as a test framework against which laboratory sheets of paper obtained by other methods were compared. The flow diagram of the process in the production laboratory sheets is shown in Fig. 1c. The mentioned process was compared with a method that does not use chemicals in the separation of impurities in the paper pulp, the P1 procedure (Fig. 1a). The intention of this process is to increase the sustainability of the deinking process, reduce the amount of chemicals used in the process (economic benefit) while obtaining a raw material that is qualitatively satisfactory. The next method, method P2, is a combination of standard INGEDE method 11 [22] and method P1 (Fig. 1b). The aim of this procedure was to see if the additional process would contribute to a higher quality of paper pulp. Higher pulp quality can contribute to the absence of a bleaching process, which ultimately results in less chemical use and reduced impact on the environment. Manual sheets are made according to standard INGEDE 1 procedure [23] and ISO 5269-2: 2004 [24] standards, in all phases from P1, P2 and P3. Procedures using Rapid Köthen sheet former.

The following methods were used for measuring the optical characteristics of laboratory handsheets: the diffuse blue reflectance factor according to ISO 2470-1:2016 [25], effective residual ink concentration, ERIC according to TAPPI T 567:2009 [26], ISO 22754:2008 [27] and color determination for paper and board, ISO 5631-3:2015 [28]. Image analysis was used to count the detected residual impurity particles and area. Spec × Scan Apogee System image analysis software according to ISO 13322-1, 2014 [29] was used, which includes a scanner to digitalize images. The values on the device were as follows: threshold value (100), white level (75) and black level (65) were chosen after comparing the computer images to the handsheets.

3 RESULTS AND DISCUSSION

By studying the results of the ISO brightness measurement of the handsheet samples obtained from printed boxes and printed media after the separation of impurity particles, it is observed that all the values increase following this procedure. Therefore, it can be concluded that all the procedures contributed to an increase in brightness and the impurity particles had been separated. The highest measured values were obtained using the INGEDE Method 11, which is the standard method for recycling paper and cardboard.

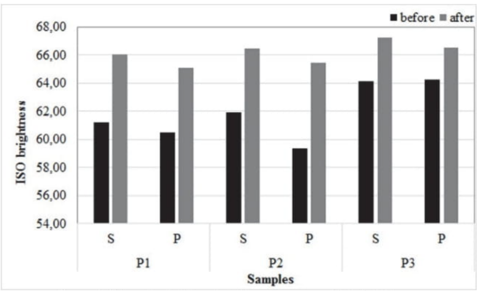

Figure 2 The effect of the deinking process on ISO brightness

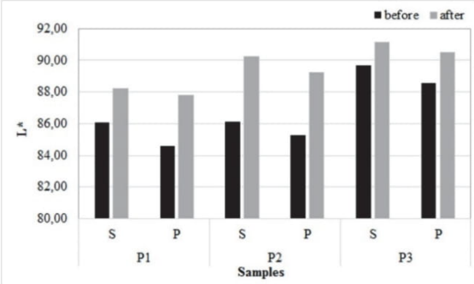

Figure 3 The effect of different impurity separation procedures on the values of the colorimetric coefficient *L**

Unlike earlier interpreted results, the values of the colorimetric coefficient *L** show the influence of the reflection of color as a combination of tint, saturation and dark/light value. *L** measures are the lightness of the samples. The trends of the measured values of the handsheets before and after the impurity separation process follow the trends measured for ISO brightness and contribute to the confirmation of the previously measured results.

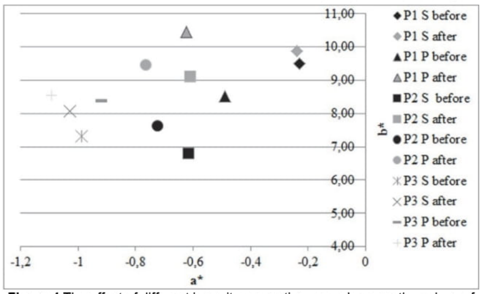

Figure 4 The effect of different impurity separation procedures on the values of the colorimetric coefficients *a** and *b**

When the values of the colorimetric coefficient *b** of the handsheets obtained from printed laminated substrates are examined, it is evident that the optical brighteners are separated after the separation of impurities because the handsheets colorimetrically go toward yellow tones. It should be explained that in the first and second decantation processes, in addition to dye particles, optical brighteners are also rinsed off.

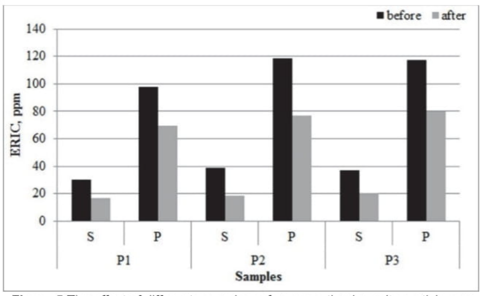

Figure 5 The effect of different procedures for separating impurity particles on the ERIC value

The same trend is also present for samples obtained from printed laminated boxes, although the value coefficient *a** decreases, which means that the sheets acquire a slightly bluish tone. The reason for this phenomenon is that the printing on the box is blue-purple. This trend is most noticeable with the standard INGEDE Method but it is important to mention that these samples lose the least optical brighteners and do not have a greatly increased *b** value.

By measuring samples of handsheets obtained from printed substrates with Procedure P2 prior to the separation of impurity particles, the highest values of particles were obtained but it can be seen that the measured values after the separation of the impurity particles were the lowest. Thus, it could be concluded that in Procedure P2, the largest quantity of particles generated are those that are best separated by the processes in the procedure. In general, it can be concluded that the quantity of particles on the handsheets obtained from printed substrates is small and they are efficiently separated by all the procedures. Good efficiency in the separation of impurity particles is also seen for printed laminated drug boxes. From the measured ERIC values, which are at most 80 ppm, it can be concluded that the obtained hand sheets are of satisfactory optical quality for the production of packaging products.

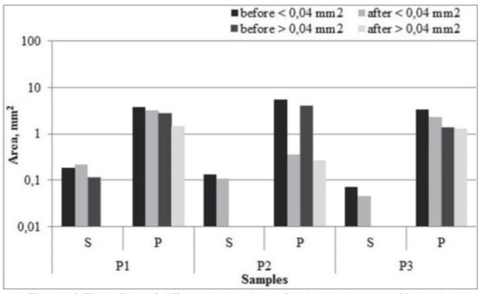

Figure 6 The effect of different procedures for the separation of impurity particles on the particle surfaces on handsheets

By studying the surfaces of the impurity particles left on the handsheets before and after the particle separation procedure for the samples of laminated print substrates, it can be established that the separation process is highly successful since, after the procedure, generally no impurity particles are presented. In Procedure 1, after the process of separating the impurity particles obtained from the substrate, the particle surfaces in the class of particles < 0,04 mm^2 increased. The reason for this phenomenon is that some of the particles become fragmented into smaller particles from the class of particles > 0,04 mm^2. It must be noted that the total surface of all the impurity particles after the particle separation process decreased by 40%. Comparing the procedures for the samples of printed laminated boxes, Procedure 2 reduces the surface area of the impurity particles of the handsheets the most after the separation of the impurity particles in both particle size classes. This method is particularly distinguished for the separation of particles belonging to the class > 0,04 mm^2. Such particles affect the inhomogeneity of the sheets of paper and are usually more difficult to separate using the conventional procedure, which can be seen in comparison to Procedure 3, where there is the least separation of such particles.

The number of particles on the handsheets after the procedure for the separation of impurity particles in both classes is generally zero. The only exception occurs in Procedure 1 in the particle size class of < 0,04 mm^2, where two particles are left. In this case, the number of particles was reduced fifteen-fold, so that even then the procedure is efficient. Given the small number of particles, it can be concluded that the particles in this class do not significantly affect the grayness of the paper. When studying handsheets made from the pulp of printed boxes before and after the separation of impurity particles, it is seen that Procedure 2 is the most efficient, where the number of particles in both classes is reduced approximately ten-fold. The remaining two procedures are equally successful in separating particles and the number of impurity particles is reduced by about half of the initial values.

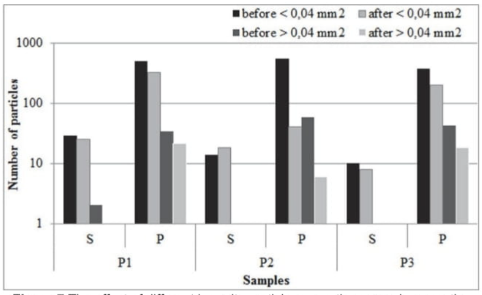

Figure 7 The effect of different impurity particle separation procedures on the number of particles on the handsheets

4 RESULTS AND DISCUSSION

Regarding the fidelity of the ISO brightness and colorimetric coefficient *L**, the greatest increase in value is seen with Procedure 2, which can be explained by the removal of the largest number of impurity particles. The same conclusion can be confirmed from the lowest ERIC values. It is nevertheless important to note that with Procedure 3, the highest ISO brightness and colorimetric coefficient CIE *L** is achieved, from which it can be concluded that this method removes the fewest optical brighteners from the samples. This fact is also confirmed by the values of the colorimetric coefficient CIE *b**, which are the lowest but higher than zero, i.e., they are in the brightest yellow range in comparison to the other samples.

By studying the number of particles on the handsheets before and after the separation of the impurity particles, it can be noted that Procedure 2 is the most successful, which is a combination of two methods for separating impurity particles. The separation performance of Procedures 1 and 3 are equal but it is significant that chemicals are not used in Procedure 1, which increases the sustainability of this method. The most successful procedure for separating impurity particles > 0.04 mm^2 is Procedure 1. This can help explain the efficiency of Procedure 2. The good fragmentation of the class of particles > 0,04 mm^2 in Procedure 1 and the presence of INGEDE deinking chemicals in Procedure 3 easily separate the formed impurity particles from paper pulp and yield the best separation results. It is important to note that by examining the optical homogeneity of the handsheets achieved by all the procedures, there are no inhomogeneities on the handsheets, according to S. Runte et al., who state that such samples have very good visual quality [30].

From this study, it can be concluded that all three methods satisfactorily remove impurity particles from pulp made from printed laminated substrates and printed laminated boxes for drugs. A specific issue in the separation of impurities from the said samples is the presence of adhesives as well as silver foil (BOPET metallized film) for lamination. There is a higher concentration of adhesive in the pulp when the adhesive is applied to the entire surface of the cardboard or the printed substrate but the adhesive used for gluing boxes should also be mentioned. All of the above contribute to the complexity of the process of separating impurity particles from paper pulp because different types of particles are formed (including stickies), many of which can agglomerate due to the presence of adhesive. The present study contributes a procedure that successfully separates impurity particles from paper pulp in a more sustainable manner, without the use of chemical agents.

5 CONCLUSION

Regarding the fidelity of the ISO brightness and colorimetric coefficient L^*, the greatest increase in value is seen with Procedure 2, which can be explained by the removal of the largest number of impurity particles. The same conclusion can be confirmed from the lowest ERIC values. It is nevertheless important to note that with Procedure 3, the highest ISO brightness and colorimetric coefficient CIE L^* is achieved, from which it can be concluded that this method removes the fewest optical brighteners from the samples. This fact is also confirmed by the values of the colorimetric coefficient CIE b^*, which are the lowest but higher than zero, i.e., they are in the brightest yellow range in comparison to the other samples. By studying the number of particles on the handsheets before and after the separation of the impurity particles, it can be noted that Procedure 2 is the most successful, which is a combination of two methods for separating impurity particles. The separation performance of Procedures 1 and 3 is equal but it is significant that chemicals are not used in Procedure 1, which increases the sustainability of this method. The most successful procedure for separating impurity particles > 0.04 mm^2 is Procedure 1. This can help explain the efficiency of Procedure 2. The good fragmentation of the class of particles > 0,04 mm^2 in Procedure 1 and the presence of INGEDE deinking chemicals in Procedure 3 easily separate the formed impurity particles from paper pulp and yield the best separation results. It is important to note that by examining the optical homogeneity of the handsheets achieved by all the procedures, there are no inhomogeneities on the handsheets, according to S. Runte et al., who state that such samples have very good visual quality [30].

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