MDF Recycling: Recovering fibres from fibreboards for further material utilisation with a focus on the chemical and morphological alteration of the recovered fibres

Dissertation

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“To live! Like a tree alone and free,

Like a forest in brotherhood

This yearning is ours!”

- Nâzım Hikmet Ran -

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Abstract

The present research was performed within the scopes of the DFG Research Training Group 1703 “Resource Efficiency in Interorganizational Networks” and AiF Project GmbH (project No. 16KN065229), which aims at developing a new fibreboard recycling technique based on thermo-hydrolytic disintegration by focusing on the further material usage of the waste fibreboards while preserving the fibrous morphology of the wood element and increasing the added-value compared to combustion (energy recovery). The results have been gathered in total as six manuscripts for being published or are submitted for publishing elsewhere.

Medium density fibreboard (MDF) is uniform, dense, smooth, and free of knots and grain patterns, and is an excellent substitute for solid wood in many applications. As the name suggests, the MDF has density range between 500 to 800 kg m^{-3} and is manufactured by hot-press consolidation with a thermo-setting adhesive resulting in an entire inter-fibre bonding of the fine lignocellulosic fibres. MDF is the second most important wood-based panel (WBP) after particleboard, hence, the global MDF production has reached above 90 million m^3 in 2017 and 2018. MDF tends to be used in indoor applications such as furniture, laminate flooring and panelling.

It is estimated that nearly 50 million m^3 of waste MDF was generated across the world in 2016 alone. Alternative approaches for the disposal of this waste are missing and need to be considered, such as recycling the waste into further value-added uses. However, a commercially viable method for MDF recycling has not been found yet. Additionally, energy recovery (combustion for energy production) is not an option in many EU countries, due to the lack of sufficient incinerator capacity for burning waste MDF. Therefore, large volumes of MDF must have accumulated across Europe. Furthermore, introducing the recovered fibres back into the MDF manufacturing as a raw material requires careful control to avoid upsetting the process or affecting the board quality.

In order to preserve the fibrous morphology of the recovered lignocellulosic fibre material, while releasing the fibres from the thermosetting resin matrix, the thermo-hydrolytic disintegration process would be the best option. Thus, in this thesis, the thermo-hydrolytic disintegration process has been chosen as the main technique for recovering the wood fibres from waste fibreboards. As a result of this disintegration process, recovered fibres (RF) and disintegration water (DW) were obtained.

Obtained recovered fibres, when compared to virgin fibres (VF), were found to be shortened which could be attributed to the disintegration conditions and have altered chemical properties resulting in higher pH and formaldehyde emissions due to the remaining resin.

Moreover, RF were further utilized for manufacturing either new MDF panels or wood polymer composites (WPC). When utilized for manufacturing new MDF, even the utilisation of 100 % RF did not cause significantly lower strength properties than that of panels made of 100 % VF. However, introducing the screw press process for drying the RF after the disintegration caused decreased strength properties compared to the original MDF panels containing VF. Although, mixing screw-pressed and air-dried RF with VF improved these properties, the strength of the panels containing solely VF were not achieved. Furthermore, WPC formulations containing RF exhibited improved mechanical and water-related properties, even though they provided similar physico-mechanical results to those containing VF.

The study has also shown that obtained DW exhibited higher pH values, N as well as formaldehyde contents while containing significant levels of reducing sugar and equivalents when compared to demineralized water.

The findings of this dissertation not only make several scientific contributions to the current literature, but also suggest utilizing RF obtained after the thermo-hydrolytic disintegration of waste MDF for manufacturing new MDF and WPC to ensure a more efficient utilization of these wood resources. Further research might be carried out to explore how different mixing proportions of RF and VF effect the strength properties of these possible utilisation areas, especially new MDF.

Zusammenfassung

Die vorliegende Studie wurde im Rahmen des DFG-Graduiertenkollegs 1703 "Ressourceneffizienz in interorganisatorischen Netzwerken" und der AiF Project GmbH (Projekt Nr. 16KN065229) durchgeführt und verfolgt das Ziel der Entwicklung eines neuen Faserplattenrecyclingverfahrens auf Basis eines thermohydrolytischen Aufschlusses. Dabei lag der Fokus auf der stofflichen Weiternutzung der Altfaserplatten bei gleichzeitiger Erhaltung der Fasermorphologie des Holzelements sowie der Erhöhung der Wertschöpfung gegenüber der Verbrennung (energetische Verwertung). Die Ergebnisse wurden in insgesamt sechs Manuskripten zusammengetragen, die zur Veröffentlichung bestimmt sind oder zur Veröffentlichung an anderer Stelle eingereicht wurden.

Mitteldichte Faserplatten (MDF) sind uniform, dicht, glatt und frei von Ästen und Maserungen und stellen in vielen Anwendungsbereichen einen leistungsfähigen Ersatz für Massivholz dar. Dem Namen entsprechend weist MDF eine Dichte zwischen 500 und 800 kg m^{-3} auf und wird durch Heißpressen mit einem wärmehärtenden Klebstoff hergestellt, welcher zu einer vollständigen Bindung der feinen Lignocellulosefasern untereinander führt. MDF ist auf dem Weltmarkt nach Spanplatten die zweitwichtigste Holzwerkstoffplatte, mit einer jährlichen Produktionsmenge von über 90 Mio. m^3 in den Jahren 2017 und 2018. MDF wird in der Regel für Innenanwendungen wie Möbel, Laminatböden und Verkleidungen verwendet.

Allein im Jahr 2016 fielen weltweit schätzungsweise fast 50 Mio. m^3 MDF-Abfälle an. Für die Entsorgung dieser Abfälle müssen alternative Ansätze, wie z.B. das Recycling der Abfälle für andere Einsatzzwecke, in Betracht gezogen werden, wobei jedoch bisher noch keine wirtschaftlich tragfähige Methode für dieses Vorhaben verfügbar ist. Darüber hinaus ist die energetische Verwertung von MDF-Abfällen (Verbrennung zur Energieerzeugung) in vielen EU-Ländern keine Option, da nicht genügend Verbrennungskapazitäten zur Verfügung stehen. Dies hat eine Ansammlung großer MDF-Mengen innerhalb Europas zur Folge. Weiterhin erfordert die Rückführung der wiedergewonnenen Fasern als Rohstoff in die MDF-Herstellung eine sorgfältige Kontrolle, damit der Prozess nicht gestört oder die Qualität der Platten beeinträchtigt wird.

Um die faserige Morphologie des zurückgewonnenen lignozellulosehaltigen Materials zu erhalten und gleichzeitig die Fasern aus der wärmehärtenden Harzmatrix zu lösen, erscheint das thermohydrolytische Aufschlussverfahren als beste Option, weshalb dieses innerhalb der vorliegenden Studie als Haupttechnik zur Rückgewinnung der Holzfasern aus Altfaserplatten gewählt wurde. Als Ergebnis dieses Aufschlussverfahrens wurden rückgewonnene Fasern (RF) und Aufschlusswasser (DW) gewonnen.

Die RF waren im Vergleich zu den Frischfasern (VF) aufgrund der Aufschlussbedingungen verkürzt und wiesen veränderte chemische Eigenschaften auf, welche anhand eines höheren pH-Werts sowie der Freisetzung von Formaldehydemissionen aufgrund des verbleibenden Harzes charakterisiert werden konnten.

Darüber hinaus wurden RF zur Herstellung von neuen MDF-Platten oder von Holz-Polymer-Verbundwerkstoffen (WPC) weiterverwendet. Bei der Herstellung ersterer führte selbst die Verwendung von 100 % RF bei Lufttrocknung nicht zu wesentlich geringeren Festigkeitseigenschaften als bei Platten aus 100 % VF. Der Einsatz des Schneckenpressverfahrens zur Trocknung der RF nach dem Aufschluss hingegen führte zu einer Verringerung der Festigkeitseigenschaften im Vergleich zu den ursprünglichen MDF-Platten mit VF. Obwohl durch die Mischung von gepressten und luftgetrockneten RF mit VF diese Eigenschaften verbessert werden konnten, wurde die Festigkeit der Platten, die nur VF enthielten, nicht erreicht. Darüber hinaus wiesen WPC-Formulierungen, die RF enthielten, bessere mechanische und wasserbezogene Eigenschaften auf, obwohl sie ähnliche physikalisch-mechanische Ergebnisse lieferten wie die Formulierungen, die VF enthielten.

Die Studie hat auch gezeigt, dass das gewonnene DW im Vergleich zu entmineralisiertem Wasser höhere pH-Werte, N- und Formaldehyd-Gehalte aufweist und gleichzeitig signifikante Mengen an reduzierenden Zuckeräquivalenten enthält.

Die Ergebnisse dieser Dissertation liefern nicht nur mehrere wissenschaftliche Beiträge zur aktuellen Literatur, sondern geben auch Anlass zu der Empfehlung, die nach dem thermohydrolytischen Aufschluss von MDF-Abfällen gewonnenen RF zur Herstellung neuer MDF und WPC zu verwenden, um eine effizientere Nutzung dieser Holzressourcen zu gewährleisten. Weitere Forschungsarbeiten könnten durchgeführt werden, um zu untersuchen, wie sich unterschiedliche Mischungsverhältnisse von RF und VF auf die Festigkeitseigenschaften dieser möglichen Verwendungsbereiche, insbesondere neuer MDF, auswirken.

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Chapter 1

Introduction

1.1 Wood-based panels (WBP)

Wood-based panels (WBP) is a general term for a variety of different board products (Figure 1.1), which have a remarkable range of engineering properties (Irle and Barbu 2010). Historically, wood was used only in its solid form as large timbers or lumbers. As the availability of large-diameter trees decreased, as well as the price increased, the wood industry looked to replace large-timber products and solid lumber with reconstituted wood products made using smaller-diameter trees and saw and pulp mill wastes (Rowell 2005). Hence, the WBP can be engineered to various specifications, thus taking the full advantage of wood's inherent properties while at the same time improving upon these properties through materials science and technology (Maloney 1996). WBP contain a significant amount of wood in assorted forms such as veneers, strands, chips, particles or fibres. Regardless of the raw material form used in the manufacture, WBP provide uniform and predictable in-service performance, largely as a consequence of standards used to monitor and control their manufacture (Cai and Ross 2010). In other words, when wood is reduced to particles or fibres and then combined in a matrix, the influence of the irregularities are reduced and strength properties exceeding those in the original state can be achieved (Rowell et al. 1993) in any desired shape and size.

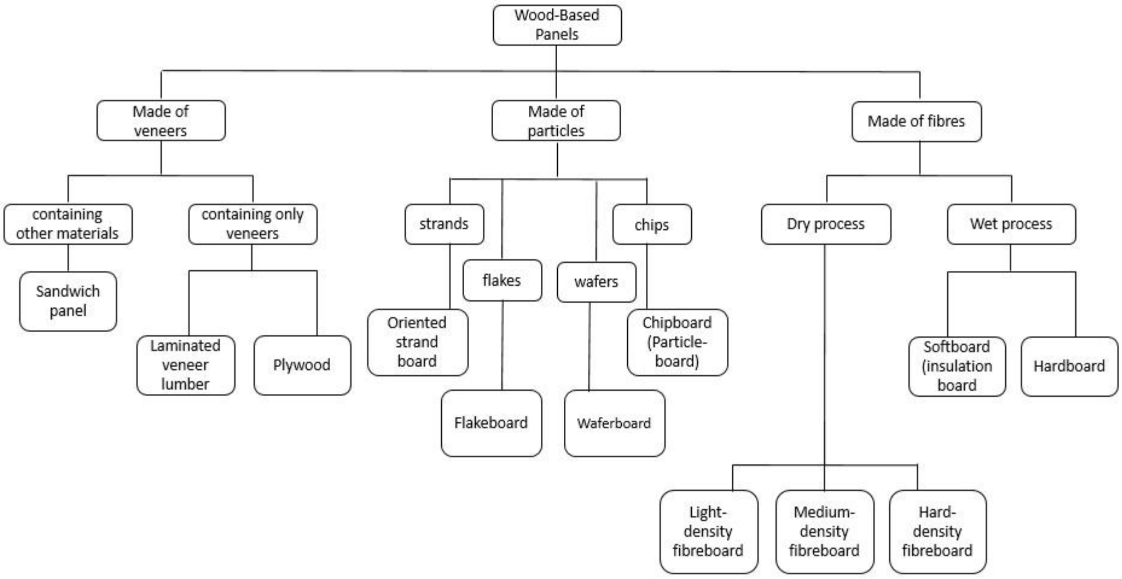

Figure 1.1: Schematic breakdown representation of wood-based panel products made of veneers, particles and fibres (adopted from (Irle and Barbu 2010)).

1.1.1 Wood fibres

Lignocellulosic fibres is a scientific name that refers to natural fibres, because all plant fibres are mainly constructed by cellulose, hemicelluloses and lignin (Hao et al. 2018), which correspond to macromolecules with chemical structures rich in hydroxyl groups (Frollini et al. 2013). In comparison to synthetic fibres, lignocellulosic fibres are biodegradable, renewable and, they can be obtained from various plants; thus they are widely available, they have low density, competitive specific mechanical properties and a relatively low cost (Panthapulakkal et al. 2006; Ching et al. 2016; Yang et al. 2019). Throughout this thesis, the term wood fibres are used as the lignocellulosic fibres. Wood is the major source of lignocellulosic fibres for producing various products, hence wood fibres comprise ca. 45 % of the total global production of the lignocellulosic fibres (Rowell 2008). In fact, wood fibre is an anatomical term and refers to a single fibre such as softwood tracheids and hardwood libriform fibres and fibre tracheids (Schirp and Stender 2010). Wood can be broken down into fibre bundles and single fibres by grinding or refining (Berglund and Rowell 2005). Today, wood fibres are predominantly obtained through a thermo-mechanical pulping (TMP) process of the wood chips using a refiner. TMP process uses the combined action of heat and mechanical energy to break the bonds between the cells that make up wood (Irle and Barbu 2010) by weakening the lignin component. During this process wood chips are continuously added to a digester and steamed at temperatures between 150 °C and 180 °C and between 3 and 5 bar pressure for several minutes, after softening, the wood is ground between two refiner discs (one rotating and the other stationary), thus fibres are extracted from the wood matrix (Schirp et al. 2014). After refining, the fibre lengths are between 1.0 mm and 5.0 mm and fibre widths are between 2.5 µm and 3.0 µm (Rowell 2008).

1.1.2 Fibreboards (FB)

Fibreboard term includes hardboard (HB), medium and high density fibreboards (MDF and HDF) and softboard (insulation board) panels. These panel products are based on wood or other lignocellulosic fibres held together by an adhesive bond, either by using the inherent adhesive properties of the wood polymers (wet-process) or by adding an adhesive (dry-process) (Rowell 2005). Therefore, FB are normally classified by density and process method (English et al. 1997). Manufacture of FB can be broken down into two main steps; disintegration of larger wood elements into fibres, and the formation of a board structure

(Berglund and Rowell 2005). Global fibreboard production reached 116 million m^3 in 2018 (FAO 2019a). Akin other WBP products, FB can be sawn, routed, spindled or drilled conveniently. Moreover, when compared to solid wood materials, FB are less expensive albeit their stable strength properties. The European Standard for wood fibreboards EN 316 (DIN EN 316 2009) defines the FB as a panel material with a nominal thickness of 1.5 mm or greater, manufactured from lignocellulosic fibres with the application of heat and/or pressure. Furthermore, FB comply with the European Standards EN 622-1 to 5 (DIN EN 622-1 2003; DIN EN 622-2 2004; DIN EN 622-3 2004; DIN EN 622-4 2009; DIN EN 622-5 2009).

1.1.2.1 Medium density fibreboards (MDF)

MDF is uniform, dense, smooth, and free of knots and grain patterns, and is an excellent substitute for solid wood in many applications (André et al. 2008). As the name suggests, the MDF has density range between 500 to 800 kg m^{-3} and is manufactured by hot-press consolidation with a thermo-setting adhesive (Park et al. 1998; Berglund and Rowell 2005) resulting in an entire inter-fibre bonding of the fine lignocellulosic fibres. MDF is the second most important WBP after particleboard, hence, the global MDF production has reached above 90 million m^3 in 2017 and 2018 (FAO 2019a, b). The first dry-process MDF factory was built in Deposit, USA, in 1966 (Maloney 1996) and the first MDF factory in Europe is thought to be that built in Ribnitz-Damgarten, Germany in 1973 (Irle and Barbu 2010). MDF manufacturing sequenced as disintegration of larger wood elements into fibres including log debarking, log chipping and screening, chip washing, chip preheating and refining, and the formation of a board structure containing resin application and blending (occurring in the blow line), fibre drying, mat forming and pre-pressing, mat hot pressing, cooling and finishing (Irle and Barbu 2010; Gonçalves et al. 2018) (Figure 1.2). The European Standard EN 622-5 (DIN EN 622-5 2009) specifies the minimum requirements for MDF panels. MDF tends to be used in indoor applications such as furniture, laminate flooring and panelling. Furthermore, MDF is now versatile as solid wood because it can be glued, stapled, and attached with dowels (Schulte and Frühwald 1996; Antov and Savov 2019; Lubis et al. 2021).

1.1.2.2 High density fibreboards (HDF)

When MDF density exceeds 800 kg m^{-3} and reaches up to 1.2 kg m^{-3}, it may also be referred to as high density fibreboard (HDF) (Berglund and Rowell 2005; Kitek Kuzman and Kutnar 2014). HDF production together with MDF accounted for 85 % of all FB production in 2018

(FAO 2019a). HDF applications include exterior siding, interior wall panelling, household and commercial furniture, and industrial and commercial products (Berglund and Rowell 2005) and foremost in laminate flooring.

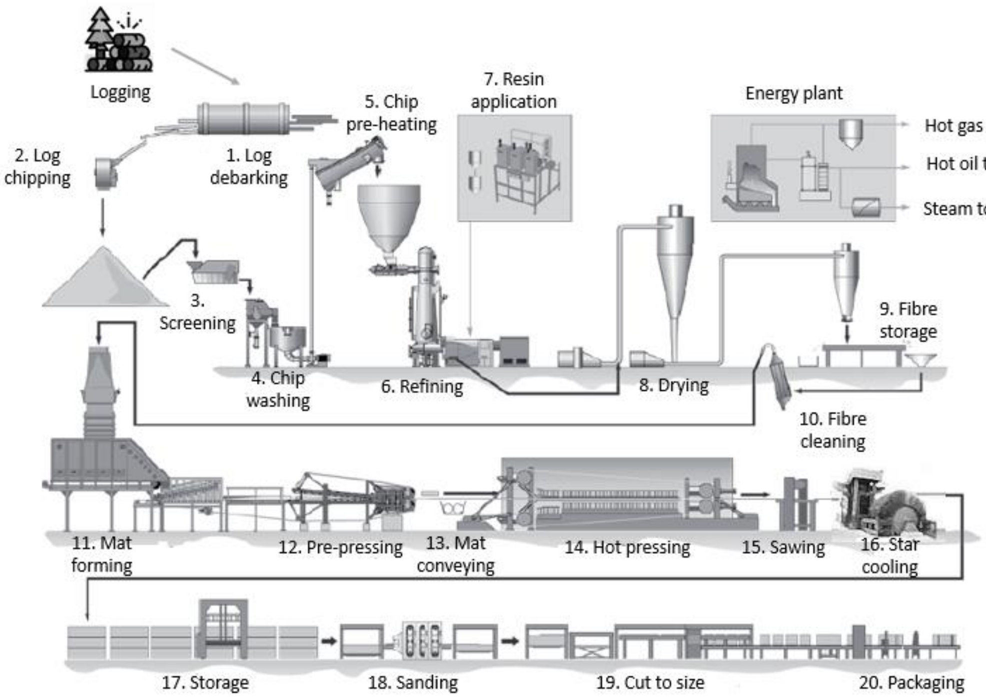

Figure 1.2: MDF manufacturing sequence (adopted and revised from (EPA 2002; Rivela et al. 2007; Irle and Barbu 2010)): 1. Log debarking, 2. Log chipping, 3. Screening, 4. Chip washing, 5. Chip pre-heating, 6. Refining, 7. Resin application, 8. Drying, 9. Fibre storage, 10. Fibre cleaning, 11. Mat forming, 12. Pre-pressing, 13. Mat conveying, 14. Hot pressing, 15. Sawing, 16. Star cooling, 17. Storage, 18. Sanding, 19. Cutting to size, 20. Packaging.

1.1.3 Wood polymer composites (WPC)

In wood polymer composites (WPC), a polymer matrix forms the continuous phase surrounding the wood component (Oksman Niska and Sain 2008). WPC are composites that exhibit thermoplastic processing behaviour and consist of varying contents of wood, (thermo)plastics and additives (e.g., coupling agent), which are processed by thermoplastic shape-forming techniques such as extrusion, injection moulding, roto-moulding or compression moulding (Vogt et al. 2006; Teuber 2016). WPCs find applications in a broad range of products, such as non-structural components like decking, fencing, industrial flooring, landscape timbers, railings, mouldings and siding as well as window and door profiles (Clemons 2002; Jeske et al. 2012) yet more in the automotive industry led by the recycling requirements by the European Commission (Ashori 2008). The wood used in WPCs is most often in particulate form (e.g., wood flour) or very short fibres, rather than longer individual wood fibres (Clemons 2002). Small fibres with high aspect ratios (width to length) are recommended for the use in WPC as they are distributed more homogeneously in the matrix than long fibres and provide a larger specific surface area, which improves the compatibility (Ashori 2008), thus act as reinforcement. Therefore, recovered fibres (RF) obtained after the thermo-hydrolytic disintegration of fibreboards might be used as a filler and/or reinforcing agent for thermoplastic polymers to produce WPC (Bütün et al. 2018). Severe fibre length reduction, however, occurs during the composite preparation because of the chosen raw material feeding method, low polymer melt flow rate and increasing screw speed in the twin screw extruder as well as high shear forces during the injection moulding process (Teuber et al. 2016a, b). Moreover, processing difficulties, such as feeding and metering low-bulk-density wood fibres, limit the use of high content WPCs due to entanglement (Ashori et al. 2011).

1.2 Adhesive systems used in MDF production

In the WBP industry a great variety of adhesives such as aminoplastic resins, phenolic resins and isocyanates are currently in use (Dunky 2003). Aminoplastic resins are the most important and used adhesives for MDF production. These resins are polymeric condensation products of the reaction of aldehydes (primarily formaldehyde) with compounds such as urea carrying amino (-NH_2) or amido groups (Pizzi and Mittal 2003; Stokke et al. 2014a; Gonçalves et al. 2018). Thermosetting urea-formaldehyde (UF) resins are a product of the reaction between urea (CH_4N_2O) and formaldehyde (CH_20) (Figure 1.3 and 1.4), moreover they are the most prominent member of aminoplastic resins (P. Singh et al. 2013; Kumar and Pizzi 2019). Hence, the adhesives used in the European MDF industry with resin addition levels of 8-10 % are mostly UF, often as melamine-fortified UF (with 1-4 % melamine), so called MUF binders (Mantanis et al. 2018). The use of UF-resins as a major adhesive in MDF industry is due to a number of advantages, including low cost, ease of use under a wide variety of curing conditions, low cure temperatures, water solubility, resistance to microorganisms and to abrasion, hardness, excellent thermal properties, and lack of colour. However, their lack of resistance to moist conditions, especially in combination with heat leads to a reversal of the bond-forming reactions and the release of formaldehyde (Conner 1996). In other words, UF-resins’ bonds deteriorate caused by liquid water and water vapour, due to the hydrolysis of aminomethylenic bond (Pizzi and Mittal 2003), which is mostly dimethylether bond.

Melamine-urea-formaldehyde (MUF) resins, on the other hand, have similar formulation to UF resins, but a certain amount of the urea is replaced by melamine in order to increase the resistance to hydrolysis. It is suitable as a replacement of UF, albeit considerably more costly, to bond interior products (Stokke et al. 2014a). Hence, there is a motto for MUF production which is “applying as much melamine as necessary, but as little as possible for economic reasons” (Gonçalves et al. 2018).

Moreover, 2 % to 6 % organic isocyanates based on dry fibre mass are used, usually polymeric diphenylmethane diisocyanate (pMDI), which is a mixture that contains 25-80 % monomeric 4,4’-MDI as well as oligomers containing 3-6 rings and other minor isomers, such as the 2,2’-isomer. pMDI adhesives can be additionally used as an accelerator and as a special linker for UF resins (Dunky 2003), thus the disadvantages of the UF resin (such as formaldehyde emission) can then be compensated and susceptibility against hydrolysis can be enhanced. However, MDF, bonded exclusively with pMDI or in combination with UF resin, comprises a very small market in Europe, constituting less than 1 % of the whole MDF production (Mantanis et al. 2018).

1.2.1 Formation of urea-formaldehyde (UF) resins

The formation of UF resins proceed in two stages (Conner 1996). In the first stage, urea is hydroxymethylolated by the addition of formaldehyde to the nitrogen containing amino groups forming mono-, di- and trimethylolureas (Figure 1.3).

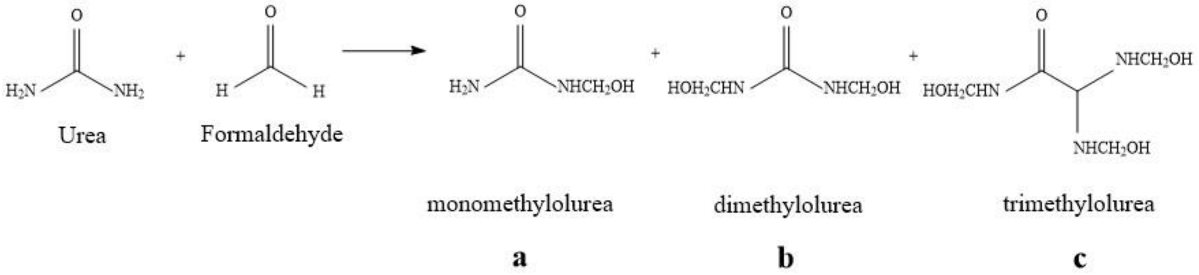

Figure 1.3: Hydroxymethylolation and formation of (a) mono-, (b) di- and (c) trimethylolurea by the addition of formaldehyde to urea (adopted from Conner 1996).

During the second stage of the UF-resin formation, a combination of (poly)condensation reactions of methylolureas occur concurrently, and eventually higher molecular weight oligomers and polymers are produced. In the first reaction, by the reaction of methylol and amino groups on reacting molecules, methylene bridges between amido nitrogens are formed (Figure 1.4a). In a side reaction, by the reaction of two methylol groups, methylene ether linkages are formed (Figure 1.4b). In another side reaction, by the splitting out of formaldehyde, methylene linkages are formed from methylene ether linkages (Figure 1.4c).

In the last side reaction, by the reaction of methylol groups splitting off water and formaldehyde, methylene linkages are formed (Figure 1.4d).

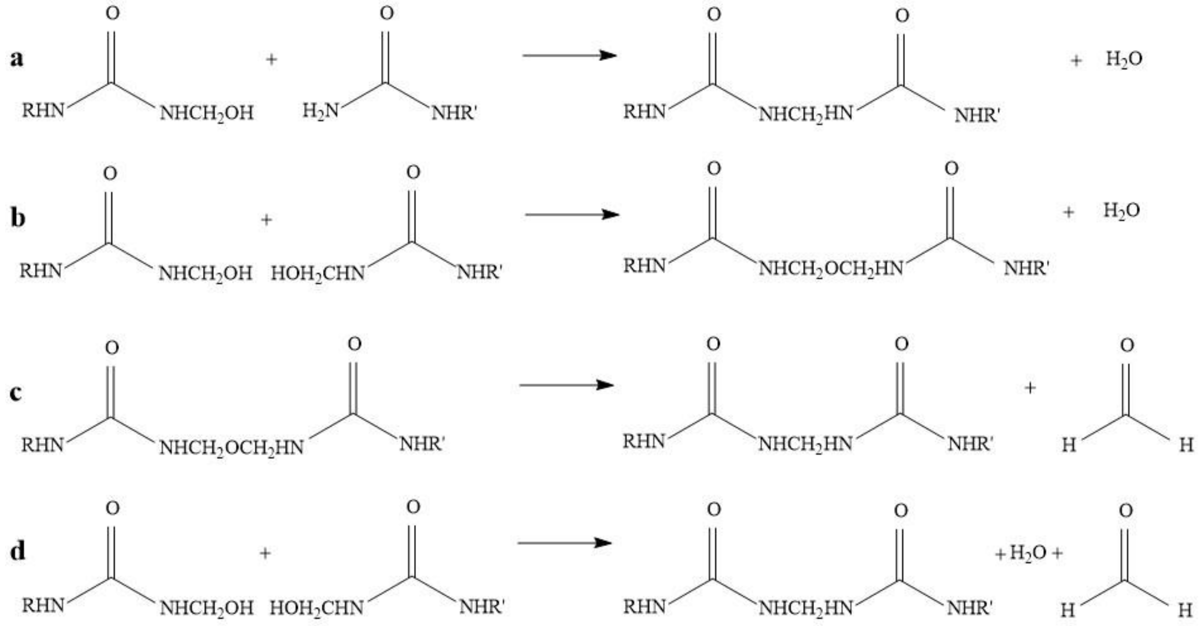

Figure 1.4: Condensation reactions of methylolureas to form (a) methylene bridges between amid nitrogens, (b) methylene ether linkages, (c) methylene linkages and (d) methylene linkages (adopted from Conner 1996).

1.2.2 Hydrolysis of urea-formaldehyde (UF) resins

As the UF-resins are the major commercial adhesive in the WBP industry, it is important to know its hydrolysis mechanism during the WBP recycling process. During the thermo-hydrolytic disintegration of WBP containing UF-resin, two hydrolysis reactions are arise; hydrolysis of the wood components and hydrolysis of UF-resin (Roffael et al. 2016; Moezzipour et al. 2017; Hagel and Saake 2020). The hardened UF-resin contains, in addition to the easily hydrolysable methylol groups, difficultly hydrolysable methyl ether linkages (Franke and Roffael 1998a). The hydrolysis of a hardened UF-resin is defined by the fragmentation of its polymer structure which takes place mainly through the cleavage of methylene (Figure 1.5a) and methylene-ether bridges (Figure 1.5b) and by the separation of amine molecule (NH_2) and the intermediate formation of carbamic acid (H_2N-COOH) (Figure 1.5c) (Fleischer and Marutzky 2000). Thus, formaldehyde and urea derivates can then be split off from hydroxymethyl groups ($-CH_2-OH$) (Hagel and Saake 2020). Furthermore, in aqueous environment, ammonia is converted to NH_4OH (ammonium hydroxide) which results in increased pH value (Moezzipour et al. 2017). Moreover, after the WBP recycling, reactivation of the used UF-resin might be observed, due to the reformation of amino and N-methylol groups as well as free formaldehyde.

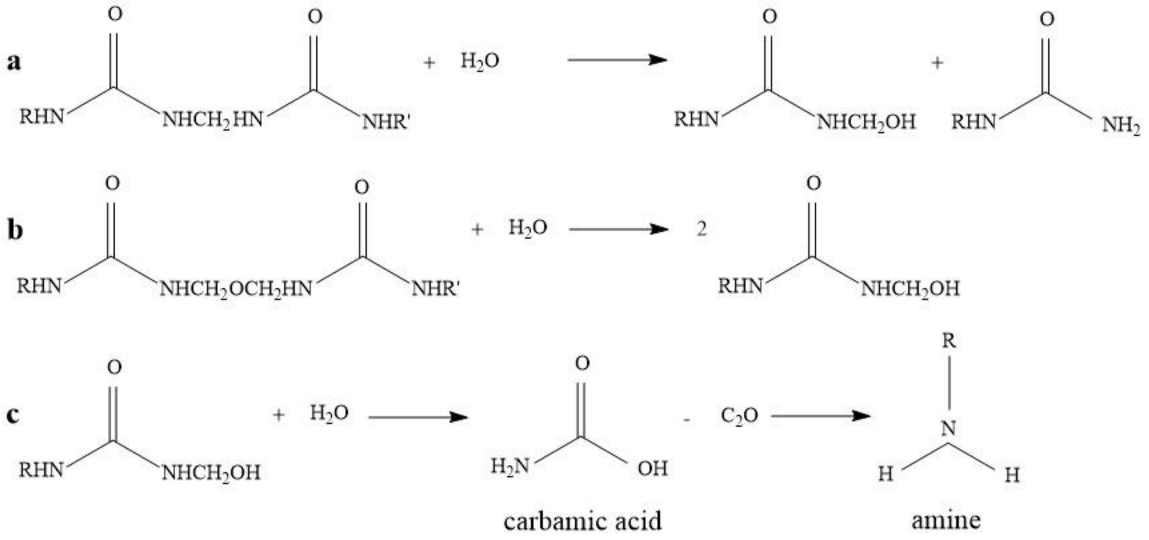

Figure 1.5: Hydrolysis of urea-formaldehyde resin. Possible degradation pathways of urea-formaldehyde resin through (a) cleavage of methylene linkages, (b) cleavage of methylene ether linkages and (c) separation of an amine molecule (NH2) and intermediate formation of carbamic acid (CH3NO2) (adopted from Fleischer and Marutzky 2000).

1.3 WBP recycling

Wood management is a key action to optimize the use of resources and to reduce the environmental impact associated with mankind activities (Rivela et al. 2007). After service life, WBP become waste and their majority will be deposited on landfills or burned. In Europe, following the environmental regulations, waste wood materials (including waste WBP) containing more than 5 % of organic materials have not been authorised for landfills since 2005 (Kharazipour and Kües 2007). The reason is, that the interaction between organic materials and the environment is of a very complex nature, leached chemicals (mainly adhesives used in the WBP manufacture) may influence the groundwater as well as the biological degradation on the landfill leads to the formation of methane , which contributes to the greenhouse effect about 80 times more than carbon dioxide (Athanassiadou et al. 2005). Even though, the word “waste” projects a perception of a material with no value or useful purpose (Rowell et al. 1991), WBP manufacturers consider recycling these materials as an alternative wood source in additional to virgin wood, which is progressively subjected to supply shortage. Thus, several processes have been developed in order to utilize waste WBP into new products. As these wastes consist to about 90 % of wood, the recovery and recycling of wood particles from waste WBP in the production of new WBP constitutes a rational mean of waste wood utilization (Lykidis and Grigoriou 2008). Furthermore, the efficient reuse and recycling of waste WBP presents an opportunity to extend wood resources, to reduce consumption of new resources, landfills, energy as well as cost through avoided purchase/disposal fees and to create “green” jobs (Bratkovich et al. 2009).

WBP containing thermosetting-resin can be recycled through applying one of the three disintegration processes, namely, mechanical, thermo-hydrolytic and chemical-assisted disintegration processes. In the mechanical disintegration process, the waste WBP are subjected to shear forces to be crushed into smaller pieces that are directly used to produce new WBP. The waste WBP are used as delivered and the disintegration proceeds in the absence of water. Therefore, the thermosetting binder is not cleaved (hydrolysed) and reactivated; fracturing of the panels mostly occurs within the wood components.

The thermo-hydrolytic disintegration process is performed for waste WBP, which are bonded with hydrolysable binders. After crushing of the material, liquid water, steam and pressure are used to cleave existing adhesive bonds in waste WBP (Kharazipour and Kües 2007). In water or under saturated steam, the bonds in the thermosetting binder are hydrolytically split, resulting in the detachment of wood particles, which may than be reused to produce new WBP.

The chemical-assisted disintegration process involves additional chemicals in order to assist the thermo-hydrolytic disintegration process by hydrolysing the binder. This process is suitable for disintegrating waste WBP containing hardly hydrolysable and non-hydrolysable binders.

1.3.1 MDF recycling

Taking into consideration that MDF can be used rougly for 30 years in decorative and interior applications before disposal (Deak 2013), it is estimated that nearly 50 million m^3 of waste MDF was generated across the world in 2016 alone (Irle et al. 2018). This estimate is based on the evolution of production capacity combined with a waste conversion model. In view of the fact that Europe produces about 18 % of the world’s MDF, combined with the fact that Europe is a net importer of furniture, which contains MDF, it is highly likely that at least 218 million m^3 of waste MDF have been created in Europe in the last five years. Moreover, a great deal of MDF waste is produced, in the form of process waste and off-cuts. There is evidence that the current disposal practices for waste MDF have significant environmental and economic concerns associated with them. Due to this, alternative approaches for the disposal of this waste need to be considered, such as recycling the waste into other uses. However, a commercially viable method for MDF recycling does not yet exist. Additionally, energy recovery (combustion for energy production) is not an option in many EU countries, due to lack of sufficient incinerator capacity for burning waste MDF (Irle et al. 2018). Therefore, large volumes of MDF must have accumulated across Europe. Furthermore, introducing the recovered fibres back into the MDF manufacturing as a raw material (Figure 1.2 after point 6 and before point 7) requires careful control to avoid upsetting the process or affecting the board quality (Smith 2004).

Recycling of waste MDF is challenging because, the fibrous morphology of the wood elements should be preserved during the recycling process. As for WBP in general, previous studies in the literature to recycle MDF involved also various thermo-hydrolytic (Sandison 2002; Jawaid 2008; Beele 2009; Roffael et al. 2010; New 2012; Moezzipour et al. 2018) and chemical (Michanickl and Boehme 2003; Nakos et al. 2005; Wan et al. 2014; Lubis et al. 2018a) processes. Moreover, Park et al. (2013) attempted to add value to MDF waste by extracting bio-oil and biogas through fast pyrolysis while Couret et al. (2017) extracted cellulose nanocrystals (CNC) by separating the lignocellulosic and thermosetting resin components of the waste MDF using alkali and bleaching treatments. Although some of these laboratory methods were promising, none of them was able to reach the next level for scaling up and meet the industry requirements, with an exception of Michanickl and Boehme’s recovery process which was in use for a short while. Limitations have to do with the complexity of methods, associated high costs and energy consumption, low quality of the RF (altered morphological and chemical properties of due to the severe disintegration conditions) and low removal levels of the cured resin (Lubis et al. 2018a). Even though, it might be advantageous considering the reactivation possibility of the resin after applying the thermo-hydrolytic process.

In order to preserve the fibrous morphology of the recovered lignocellulosic fibre material, while releasing the fibres from the thermosetting resin matrix, the thermo-hydrolytic disintegration process would be the best option. Thus, in this thesis, the thermo-hydrolytic disintegration process has been chosen as the main technique for recovering the wood fibres from waste fibreboards.

1.4 Terminology

The terms reuse, reutilize, recycle, recover, etc. are often used interchangeably among experts and in the literature. According to European Commission (2014), reuse of waste means any operation by which products or components that are not waste are used again for the same purpose for which they were conceived. Recycling of waste is defined as any recovery operation by which waste are processed into products, materials or substances whether for the original or other purposes. Recovery of waste means any operation the principal result of which is waste serving a useful purpose by replacing other materials which would otherwise have been used to fulfil a particular function, or waste being prepared to fulfil that function, in the plant or the wider economy. Through recovery, wastes are used as an input material to create valuable products as new outputs. Throughout this thesis, the term recovered is used to identify the obtained wood fibres after conducting the thermo-hydrolytic disintegration of the fibreboards. Moreover, the term recycled is used to identify the manufactured MDF panels containing either fully (100 %) or partially (50 %) recovered fibres (RF).

1.5 Objectives and outline of the thesis

The present research was performed within the scopes of the DFG Research Training Group 1703 “Resource Efficiency in Interorganizational Networks” and AiF Project GmbH (project No. 16KN065229), which aims at developing a new fibreboard recycling technique based on thermo-hydrolytic disintegration by focusing on the further material usage of the waste fibreboards while preserving the fibrous morphology of the wood element and increasing the added-value compared to combustion (energy recovery). Therefore, the following research questions are investigated:

- What are the available disintegration processes for waste WBP?
- What are the optimal thermo-hydrolytic disintegration conditions for disintegrating waste MDF?
- Is the utilization of RF obtained after the thermo-hydrolytic disintegration of the waste MDF suitable for new MDF and WPC production?

- How coherent is to utilize RF obtained after the thermo-hydrolytic disintegration of the waste MDF in manufacturing new MDF panels?
- How does the thermo-hydrolytic disintegration process of the waste MDF affect the chemical and morphological properties of the fibre material?
- What is the chemical composition of the disintegration water (DW) obtained after the thermo-hydrolytic disintegration of the waste MDF?
- How do the different proportions of RF affect the mechanical and physical properties of WPC containing RF?
- What effects do RF obtained by different waste MDF disintegration processes have on the properties of WPC?

Therefore, Chapter 2 of the thesis reviews the processes that are available for disintegrating waste WBP while serving as an extended introduction chapter. Moreover, it discusses the studies with emphasis on the production of new WBP (such as particleboards, MDF and WPC) from the recycled WBP.

Chapter 3 focuses on the repeated thermo-hydrolytic disintegration of MDF and aims to put back obtained RF into the MDF manufacturing process as closed-loop recycling. Apart from the assessment of the recycled MDF panels’ mechanical and physical properties, chemical properties of the obtained RF and disintegration water after the disintegration processes are also investigated.

Chapter 4 characterises recovered fibres’ morphology obtained after thermo-hydrolytic disintegration of MDF and compares them with the virgin fibres’ morphology by using three fibre analysers.

Chapter 5 presents a drainage process of RF using a screw press after thermo-hydrolytic disintegration of waste fibreboards. Besides, it attempts to reutilize the obtained RF in new MDF production.

In Chapter 6, mechanical and physical properties of WPC containing RF are evaluated and compared to identically produced composites containing virgin fibres.

Similarly, Chapter 7 engages in introducing milled RF obtained by thermo-hydrolytic and mechanical disintegration of waste MDF in WPC production while evaluating the mechanical and water-related properties.

Lastly, Chapter 8 provides a comprehensive interpretation, thus discussion of the results obtained in the previous six chapters and concludes with a summary of them.

1.6 Authorship

The presented studies in Chapters 2 to 7 are a reproduction of manuscripts (either published or intended to be published elsewhere), they were created in a collaboration with co-authors. Therefore, at the beginning of the each chapter, the contribution of each co-author is evaluated according to the authorship index of Hunt (1991). Moreover, a detailed corresponding description of each co-author’s contribution is also presented.

Chapter 2

Utilization of waste wood-based panels for the production of new ones with emphasis on particleboards, medium density fibreboards (MDF) and wood polymer composites (WPC): A review

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Authorship

Table 2.0: Contribution of the co-authors (as modified from Hunt 1991).

	Planning & Design	Executing & Data Collection	Data analysis & Interpretation	Literary input	Σ
F. Yağmur Bütün Buschalsky	20	25	25	25	**95**
Stergios Adamopoulos	15	5	15	5	**40**
Carsten Mai	15	0	15	15	**45**

F. Yağmur Bütün Buschalsky defined the scope of the literature review together with Stergios Adamopoulos and Carsten Mai. Moreover, she analysed and interpreted the literature and wrote the manuscript.

Stergios Adamopoulos defined the scope of the literature review together with F. Yağmur Bütün Buschalsky and Carsten Mai. He supported the literature interpretation and manuscript writing.

Carsten Mai defined the scope of the literature review together with F. Yağmur Bütün Buschalsky and Stergios Adamopoulos. He assisted the literature interpretation and manuscript writing.

2.1 Abstract

Global wood-based panels’ (WBP) production (including particleboards, oriented strand boards, plywood and fibreboards) totalled 407.5 million m^3 in 2018. Eventually, these panels will be converted to waste after their service-life. Due to the foreseen shortage of raw material supply, WBP manufacturers are looking for ways to broaden the supply of raw material sources and the utilization of waste wood as one option. To utilise recovered wood particles and fibres obtained from waste WBP recycling can provide a favourable raw material for producing new WBP and help to develop a circular bioeconomy in this field. Several processes have been developed on a pilot or industrial scale in order to turn waste WBP into new value-added products. These processes include mechanical, thermo-hydrolytic and chemical-assisted disintegration techniques, which are described in a large volume of published studies or filed as patents. This paper reviews these studies with emphasis on the production of new particleboards, medium density fibreboards and wood polymer composites from the recycled WBP.

2.2 Keywords

Wood-based panels; waste; recycling; fibreboard; MDF; particleboard; wood-polymer composites

2.3 Introduction

Wood-based panels (WBP) is a generic term for a variety of different board products made of fibres, particles or veneers (Thoemen et al. 2010) to be used for furniture and construction purposes. Global WBP production (including particleboard - PB, oriented strand board - OSB, plywood – PW, and fibreboard - FB) totalled 407.5 million m^3 in 2018 (FAO 2019a). After service life, these panels will become waste and their majority will be deposited on landfills or burned. In Europe, waste wood materials containing more than 5 % of organic materials have not been authorised for landfill since 2005 (Kharazipour and Kües 2007). This can therefore because of the fact that the disposal of these wastes may be a source of formaldehyde emission due to the degradation of urea-formaldehyde resins by microorganisms (Lubis et al. 2021). Even though, the word “waste” projects a perception of a material with no value or useful purpose (Rowell et al. 1991), WBP manufacturers consider recycling these materials as an alternative wood source in additional to virgin wood, which is progressively subjected to supply shortage. Thus, several processes have been developed in order to utilize waste WBP into new products. As these wastes consist of about 90 % of wood, the recovery and recycling of wood particles from waste WBP in the production of new WBP constitutes a rational mean of waste wood utilization (Lykidis and Grigoriou 2008). Furthermore, the efficient reuse and recycling of waste WBP presents an opportunity to extend wood resources, to reduce consumption of new resources, landfills, energy as well as cost through avoided purchase/disposal fees and to create “green” jobs (Bratkovich et al. 2009). Thus, the utilization of waste WBP can maintain the absorbed carbon inside the wood products for long periods (Kim and Song 2014). In addition, one of the main restrictions for WBP manufacturers and waste management companies is related to high NO_x emissions created when amino resin-containing waste WBP are burned in conventional combustion systems (Smith 2004). Recycling of waste WBP is an opportunity to prevent these emissions but restricts the possibility to gain thermal energy by combustion.

Waste wood such as solid wood, plywood, laminated veneer lumber (LVL) and PB may be re-used after mechanical disintegration under shear forces to produce new PB; the particleboard industry quickly adopted recycling in the 1990s (Earl and Elias 1997; Elias and Bartlett 2018). In contrast, the only currently available method to manage waste medium density fibreboards (MDF) and other WBP types is burning or landfilling (Irle et al. 2018). The main source for the wood component in wood polymer composites (WPC) is wood flour, the primary side product of sawmills (Gozdecki et al. 2015).

The objective of this review is to shed light on the following questions:

- What type of processes are available for disintegrating waste WBP (including PB, OSB, PW and MDF)?
- Which are the applications for disintegrated waste WBP?
- What are the benefits and/or consequences of utilizing disintegrated waste WBP for producing new WBP with emphasis on PB, MDF and WPC?

2.4 Overview of waste WBP disintegration processes

This chapter reviews the patents for waste WBP disintegration (Table 2.1). So far, three types of processes have been developed for disintegrating thermosetting binder-containing waste WBP. These include mechanical, thermo-hydrolytic and chemical-assisted disintegration processes. In the mechanical disintegration process, the waste WBP are subjected to shear forces to be crushed into smaller pieces that are directly used to produce new WBP. The waste WBP are used as delivered and the disintegration proceeds in the absence of water. Therefore, the thermosetting binder is not cleaved (hydrolysed) and reactivated; fracturing of the panels mostly occurs within the wood components.

Table 2.1: Patented waste WBP disintegration processes.

Reference	Type of disintegrated waste WBP	Type of targeted end-product	Disintegration method
(Kharazipour and Nonninger 2001)	PB, MDF	PB	Mechanical
(Sandberg 1965)	PB, FB	PB	Thermo-hydrolytic
(Roffael et al. 1996)	PB, FB	PB, FB	Thermo-hydrolytic
(Pfleiderer GmbH & Co. KG 1994, 1995, 1999)	PB, FB, PW	PB, FB	Thermo-hydrolytic
(Sandison 2002)	PB, FB	PB	Thermo-hydrolytic
(Kharazipour et al. 2002, 2004a, b)	PB, MDF	FB	Thermo-hydrolytic
(Jawaid 2008)	PB, FB, OSB	FB	Thermo-hydro-electrical
(New 2011, 2012)	FB	FB	Thermo-hydro-electrical
(Elias et al. 2012)	MDF	MDF	Thermo-hydro-electrical
(Michanickl and Boehme 1995, 1996, 1998)	PB, FB	PB, MDF	Chemical-assisted
(Roffael and Dix 1998)	PB, FB	-	Chemical-assisted
(Roffael 1998, 2002)	PB, FB	PB, FB	Chemical-assisted
(Nakos et al. 2001, 2005)	PB, FB, OSB	FB	Chemical-assisted

The thermo-hydrolytic disintegration process is performed for waste WBP, which are bonded with hydrolysable binders (such as urea formaldehyde - UF). After crushing of the material, liquid water, steam and pressure are used to cleave existing adhesive bonds in waste WBP (Kharazipour and Kües 2007). In water or under saturated steam, the bonds in the UF binder are hydrolytically split, resulting in the detachment of wood particles, which may then be reused to produce new WBP. Hydrolysis leads to reformation of amino and N-methylol groups as well as free formaldehyde and may thus reactivate the UF binder.

The chemical-assisted disintegration process involves additional chemicals in order to assist the thermo-hydrolytic disintegration process by hydrolysing the adhesive. This process is suitable for disintegrating waste WBP bonded with hardly hydrolysable and non-hydrolysable binders (such as melamine-urea-formaldehyde - MUF and isocyanates - pMDI).

2.4.1 Mechanical disintegration processes

Mechanical disintegration is the state-of-art for waste PB recycling. Although severe fracturing occurs during mechanical disintegration, a significant proportion of recovered particles still preserves their particulate morphology. After grinding, appropriate particles are screened and re-used in the core or surface layers together with virgin particles for producing new PB panels. Kharazipour and Nonninger (2001) described a process solely based on mechanical disintegration of amino binder-containing (mainly UF) PB for the production of new WBP. The process involves disintegration of the panels in a crusher or chipper and further shredding in a hammer-mill or drum-mill. Reportedly, new PB can be produced with a content of up to 50 % of the recovered particles (size of 0.1 to 40 mm) from residual materials (including sanding and trimming) or from waste WBP after applying fresh binder and mixing with virgin wood particles. One of the main advantage of this technique is the energy saving due to the dry processes. However, the performance of the recovered material obtained by this technique is inferior compared to virgin material as a result of their lower wettability and altered morphology. Moreover, an amply amount of fine material which cannot be used in further material production is emerged.

2.4.2 Thermo-hydrolytic disintegration processes

One drawback of mechanical disintegration methods is that additional items such as coatings or decorative surface laminates of the panels are difficult to separate from the wood particles and may contaminate the potential recovered product (Sandison 2002). In case of waste FB, they may negatively affect the fibrous morphology of the recovered material, due to length reduction. During a thermo-hydrolytic disintegration process not only the binders among the particles are cleaved but also the coatings are easily removed without excessive fragmentation. The very first known method of disintegrating particle-based panels to produce new PB by using a similar thermosetting binder and pressing processes was developed by Sandberg (Sandberg 1965). The process was carried out in a steam chamber under pressure between 0.1-0.5 MPa for 0.5 to 4 h aiming to hydrolyse the binder and to disengage the wood particles by means of water vapour. The author recommended adding up to 30 % of the obtained recovered particles to the core layer of the PB and using only virgin particles in the surface layers in order to reach acceptable board properties.

A process for the production of tannin-bonded PB and FB from respective waste panels was developed by Roffael et al. (1996). This method combines the binding action of condensed tannins and inherent, reactivated UF-based binder of the waste WBP. Accordingly, waste WBP containing UF binder were mechanically shredded using a cross-beater resulting in smaller particles. After applying steam injection, these particles were disengaged and blended with a 40 % tannin extract and then pressed into new WBP.

The company Pfleiderer GmbH & Co. KG developed another thermo-hydrolytic disintegration process for recycling panel materials (generally in the form of waste PB, FB and plywood bonded with UF binders with or without a surface laminate). In a pre-treatment step, the panels were mechanically shredded into small fragments (5-10.0 x 5-10.0 cm). In a thermo-hydrolytic disintegration step, the fragments were then subjected to a fixed digestion period in a saturated water vapour atmosphere at high pressure (0.3 to 0.6 MPa) and at high temperature (140-160°C) for 3 min. Lastly, the material, which had then already disintegrated into fibrous, particle-like, plate-like or similar basic constituents is then glued and pressed to form new WBP (Pfleiderer GmbH & Co. KG 1999, 1995 and 1994).

Sandison’s Fibresolve process (2002) for the recovery of wood fibres and particles from waste FB comprises a steam-hydrolytic step and a fibre/particle separation step. Waste FB scraps are admitted into a container, thereby immediately contacting with steam below atmospheric pressure for a period greater than 30 min. The subsequent screening step serves for the separation of not disintegrated residues from the disintegrated wood fibres and particles using a rotary drum.

Kharazipour et al. (2002, 2004a, b) developed a fully continuous process device for disaggregating waste WBP. This device allowed conveying the shredded material (particle size of at least 50 x 50 mm) continuously through the end-face output openings of at least three heated pressure-tight plug screws in succession, being subjected to shear forces and frictional forces occurring in the packing screws. Therein, the steam is injected under pressure (0.2-1.1 MPa) into the middle plug screw. In contrast to a conventional batch process e.g. in a closed container, this continuous process offers a substantial shortening of the thermo-hydrolytic disintegration time.

Moreover, a study by Franke and Roffael (Franke and Roffael 1998b) investigated the steam hydrolysis resistance of UF-bonded waste PB and waste MDF towards thermo-hydrolytic disintegration at 200 °C for 2, 4 and 6 h in an autoclave and the solid residue was homogenised with an Ultra-Turrax. The yield of fibres from MDF was higher than the particle yield. The authors attributed this to the fact that hydrolysable wood constituents were washed during the refining process to produce fibres out prior to MDF production. Accordingly, the pentosane content decreased stronger after hydrolysis of the PB than of the MDF. The hydrolytic treatment significantly reduced the nitrogen content of the particles and fibres by more than 50 %, while 2 h treatment time provided the lowest nitrogen values; these were progressively slightly higher after 4 and 6 h hydrolysis. The authors attributed this to a possible re-condensation of the hydrolysis products. The pH value of the recovered particle and fibre material was around 8, while that of the hydrolysed wood material without binder was around 5. This increase in pH was explained with the formation of ammonia from urea, but the authors also pointed out that hydrolysis of wood without binder results in a lower pH (mostly due to acetic acid formation).

A follow up study (Franke and Roffael 1998a) investigated the influence of other thermo-hydrolytic disintegration temperatures (140 and 160 °C) for 1, 3 and 6 h in an autoclave on the obtained recycled particles and fibres. These were compared with thermo-hydrolytically-treated reference pine wood particles on the formaldehyde emission by flask method (DIN EN 717-3 1996) and ammonia emission assessed in the flask, too. Reference pine wood particles showed increased formaldehyde emission with increasing treatment temperature and duration. Still, these increments were very low compared to emissions of recycled particles and fibres, as the reference materials did not contain UF resin. The emissions were higher for fibres from waste MDF than for particles from waste PB and decreased with treatment duration and temperature. While reference particles did not emit detectable quantities of ammonia, recycled particles and fibres released increasing amounts of ammonia with increased temperature and treatment duration, which resulted in increasing pH value in the absorption water.

Fleischer and Marutzky (2000) studied the mechanism of resin hydrolysis during a thermo-hydrolytic disintegration process of UF-bonded PB and compared the formaldehyde release with that of hardened UF-resin during thermo-hydrolytic disintegration. The results indicated that the hydrolysis of UF-resins in the panels proceeds faster than that of solely hardened UF. The authors attributed this finding to a finer distribution of the UF resin on the particles and to a lower degree of cross-linking via methylene groups in the panels. In addition, they found that a panel with a UF resin with a higher U:F ratio (1:1.5) released higher amounts of formaldehyde than one with a lower U:F ratio of 1:1, but the percentage release related to the total release was lower from the panel with a higher U:F ratio of 1:1.5, due to a higher degree of cross-linking.

Similarly, Kraft and Roffael (Kraft and Roffael 2003) investigated the thermo-hydrolytic breakdown of small pieces of UF-bonded waste MDF at four different temperatures (100, 110, 120 and 130 °C) in an autoclave with an MDF:water ratio of 1:6. Prior to hydrolysis the MDF pieces were soaked (impregnated) with water at 100 mbar vacuum. After 1 h disintegration at 100 °C, nitrogen content assessed by the Kjeldahl method and formaldehyde content revealed that about half of the used UF binder remained on the fibres, while after 3 h only about 25 % UF remained. The U:F ratio of the obtained recovered fibres was somewhat changed. Thermo-hydrolysis also increased the formaldehyde emission of the recovered fibres compared to that of the original MDF.

Roffael et al. (2009) disintegrated UF-bonded MDF and pine wood chips by thermo-mechanical pulping (TMP) and chemo-thermo-mechanical pulping (CTMP). The TMP process involved pre-treatment with steam at 170 °C and subsequent defibrillation (pulping) in a laboratory single disc refiner at 170 °C. In the CTMP process, 0.25 % NaOH (based on dry wood) was added to the pine chips during the pre-treatment. To disintegrate the MDF, the authors used either solely 0.15 % sodium hydroxide (NaOH) or a mixture of 1 % sodium sulphite (Na_2S) and 0.25 % sodium hydroxide. TMP fibres from MDF had a lower water extractive content and lower pH-value than respective CTMP fibres. The content in the cold-water extracts and the emissions of acetate and formate from TMP fibres deriving from MDF were up to about 10 times higher than from pine wood TMP. CTMP with NaOH significantly increased the content of formate and acetate in the extracts and the emission of these acids but CTMP with a mixture of NaOH and Na_2S lead to lower content and emission of formate and acetate - in case of fibres from MDF also compared to TMP. The formaldehyde emission determined by the flask method was highest from TMP fibres of MDF, followed by respective

CTMP fibres produced with NaOH alone and lowest with CTMP fibres produced with NaOH and Na_2S.

Jawaid (2008) described a thermo-hydrolytic disintegration process assisted by electromagnetic radiation (thermo-hydro-electrical). This method combines immersing of the waste material in water and subjecting to electromagnetic radiation (having a frequency in the range 10 MHz-300 GHz) for 10-25 min to heat up the water to temperatures of 60 to 90 °C. The advantage of using microwave radiation compared to conventional treatment, e.g. with steam, is a more homogeneous heating and a faster hydrolysis of the binder. After hydrolysis, the fibres are further disintegrated through mechanical agitation in water to produce fibrous suspension from which lignocellulose may be recovered by drying the suspension.

Similarly, New (2011, 2012) applied a thermo-hydro-electrical process to disintegrate waste FB and to re-use the recovered fibres for substituting for virgin wood fibre. The method comprises two main steps; the first step is mixing shredded FB pieces with water at a temperature between 80 to 99 °C to form a slurry, and the second step is passing an electric current through the slurry to heat (up to 160 °C) the water containing the FB pieces in a closed chamber. Due to water evaporation, the pressure rises above atmospheric pressure and cause rapid hydrolysis of the binder.

The BioComposite Centre, Bangor, UK together with the company MDF Recovery Ltd. developed another thermo-hydro-electrical process for disintegrating waste MDF. This process exhibits high technological readiness to recover fibres from FB. Pre-shredded MDF fragments are soaked in an aqueous medium and then heated to around 90 °C using microwave heating to hydrolyse the binder, disintegrate the panel fragments and detach laminates adhered to the surfaces, leaving a mush that fluffs up after drying (Elias et al. 2012).

2.4.3 Chemical-assisted disintegration processes

The aim of chemical-assisted disintegration processes is to cleave bonds in binders that are less readily hydrolysable as UF resins such as melamine-urea-formaldehyde (MUF). Michanickl and Boehme (1995, 1996 and 1998) reported on a thermo-hydro-chemical method for PB and FB that comprises a step of impregnating the waste WBP with an alkaline solution consisting of water, urea and soda (sodium carbonate) lye and a subsequent heating step to

80-120 °C. Impregnation of the pre-crushed waste materials by immersing in the solution in a static or rotating pressure vessel allows swelling until they have absorbed at least 50 % of their own weight. The impregnated material is then heated to a temperature between 80 and 120°C, which leads to cleavage of the binder and disintegration of the panel debris. Hereafter, the disintegrated material is separated from other components by one or more processes such as sieving and wind screening. The impregnation solution contains soda lye to establish a high pH value and catalyse hydrolysis. Urea added can scavenge formaldehyde released from the binder (e.g. from MUF). The added chemicals may improve the quality of the obtained recovered particles/fibres because they help to prevent their decomposition.

Roffael and Dix (1998) described a procedure, in which shredded waste PB and FB were thermo-chemically pulped in an alkaline medium (18 % NaOH). The pulping process dissolves the wood structure into its main components - cellulose, hemicellulose and lignin. After pulping, the liquid remaining (waste liquor) can be separated from the cellulose and used as an extender for wood binders.

A chemically assisted thermo-hydrolytic disintegration process for producing fibrous or particulate material from waste PB or FB was developed by Roffael (1998 and 2002). This process follows the principle of alkaline sulphate (kraft) pulping, which is normally used to remove lignin and to produce chemical pulp for paper making. After shredding of the panels to smaller pieces, these are subjected to liquid water or steam at 40-12 0°C in the presence of 0.01-0.4 % by weight sodium sulphide (Na_2S) and 0.01-0.4 % by weight sodium hydroxide (NaOH). Mechanical disintegration and de-watering simultaneously or subsequently proceed due to a high shear treatment in a twin-screw extruder (TSE). The fibres and particles obtained in this process exhibit a reduced content of hemicelluloses and lignin; consequently, the fibres contain higher proportions of cellulose. The inventor suggests the application of these fibres to produce new FB.

Nakos et al. (2001 and 2005) developed another chemically assisted disintegration process for producing new FB from waste PB and FB. This process resembles chemo-thermo-mechanical pulping (CTMP), which is used to produce pulp for paper making. The chemical pre-treatment proceeds via alkaline sulphite pulping. After size reduction, the shredded waste WBP are pre-treated with an aqueous solution of sodium sulphite (Na_2SO_3) in a pre-heater unit of a refiner system under overpressure due to the formation of steam.

The treatments leads to disruption of adhesive bonds, cleavage and solubilisation of lignin and hydrolysis and degradation of hemicelluloses in the wood fibres (Roffael and Dix 1998). After pre-treatment, the panel fragments are disintegrated into fibres in a disc refiner unit of the refiner system. The separated fibres are further processed to form fibreboards in a dry process (over 5 % by weight of a feed of virgin wood material). The presence of residual adhesive after disintegration may reduce the binder consumption due to the reactivation of the binder in waste WBP.

2.5 Challenges in waste WBP recycling

PB manufacturers are the main users of waste wood, which is currently recycled by chipping or grinding. The presence of contaminants, i.e. the non-wood components, causes a range of technical problems while the inclusion of MDF and other FB in the waste wood stream is problematic because their breakdown forms low quality particles. In detail, their rough surfaces cause higher adhesive adsorption and their poor aspect ratio causes weak points in the panel. Consequently, the PB industry can tolerate as much as 5 % MDF in the supplied material and even so they had to increase the density of their products to compensate for the presence of MDF in the particle furnish. Up to now, there are hardly any recycling strategies for MDF or sorting technologies to separate them out of the waste wood streams. The traditional recycling method of other waste wood fractions (solid wood, PB, PL, etc.) is a mechanical one during which the waste material is re-chipped and new recovered wood chips are produced. The method is applied only by the PB industry and has many drawbacks, e.g. requires heavy equipment and high amount of energy, the quality of new wood chips is inferior to the initial chips, and the high amount of wood dust produced has to be rejected. The PB industry is reluctant to use more than 15 % of recovered chips in manufacturing of new PB.

The exponential growth of MDF production combined with a relatively short service life of between 14 and 20 years (Beele 2009; Irle et al. 2018) has caused an equally rapid expansion in the quantity of waste MDF. Unfortunately, a commercially viable method of recycling MDF does not exist. However, it is worth mentioning that very recently a Belgium based company UNILIN has announced their aim towards using up to 25 % recycled fibres obtained by their new recycling technology based on thermo-hydrolytic disintegration of the waste MDF panels in their MDF production by 2030. Additionally, energy recovery is not an option in many EU countries because there is not enough incinerator capacity to burn waste MDF. Therefore, large volumes of MDF must have accumulated across Europe. It is estimated that nearly 50 million m^3 of waste MDF was generated across the world in 2016 alone (Irle et al. 2018). This estimate is based on the evolution of production capacity combined with a waste conversion model. In view of the fact that Europe produces about 18 % of the world’s MDF, combined with the fact that Europe is a net importer of furniture, which contains MDF, it is highly likely that at least 218 million m^3 of MDF waste have been created in Europe in the last five years. This has caught the recycling sector off-guard. As a result, there is a high need for recycling of MDF.

Recycling waste MDF is difficult because a significant amount of urea formaldehyde (UF) resin, which is commonly used to bond wood fibres together, needs to be removed from the waste MDF. This thermosetting resin after curing becomes hard and brittle and there is no possibility of re-activate adhesion properties and processing of resin. To release fibres from the resin matrix, resin bindings are broken under the action of heat and moisture. Previous studies in the literature to recycle MDF involved various mechanical, chemical (e.g. sodium sulphite and sodium hydroxide could re-activate the cured resin and thus less resin was required in the recycled MDF), hydrolytic treatments and their combinations by applying steam and high temperature (Riddiough 2002; Sandison 2002; Michanickl and Boehme 2003; Kearley and Goroyias 2004; Nakos et al. 2005; Jawaid 2008; Beele 2009; New 2012; Wan et al. 2014; Moezzipour et al. 2018). Others attempted to add value to MDF waste by extracting bio-oil and biogas using pyrolysis (Park et al. 2013) or high value chemical structures (cellulose nanocrystals) from post-consumer MDF by the sequential fractionation of the wood constituents of MDF waste (Couret et al. 2017). Although some of these laboratory methods were promising none of them was able to reach the next level for scaling up and meet the industry requirements. Limitations have to do with the complexity of methods, associated high costs and energy consumption, low removal levels of the cured resin, and low quality of the recovered fibres (e.g. degradation due to heating and mechanical defibrillation by the refiner) (Lubis et al. 2018a). For example, the proposed thermo-hydrolytic methods in the literature require installations that consume high amount of energy while the treatment procedure is not continuous due to successive filling and discharging of the treatment vessel (Lykidis and Grigoriou 2008). Also, thermomechanical pulps from waste panels are of shorter fibre length and with higher content of fine fraction, and can be barely used even in small amounts as a partial substitute for pulps from virgin wood in making new MDF without deteriorating the physical-mechanical board properties (Roffael et al. 2010). The microrelease process that is based on dielectric heating (Mitchell and Stevens 2009; New 2012) has been demonstrated several times over the last 10 years, but has yet to achieve commercialisation. In conclusion, processing waste MDF has traditionally focussed on producing low value products for re-use, such as back in MDF. A different approach for the future could promote extracting high-value products to off-set the investment, processing and cleaning costs of waste MDF.

2.6 Utilisation of waste WBP with a focus on different end-products

This chapter reviews studies published on the utilization of waste WBP with emphasis on producing new PB, MDF and WPC (Table 2.2).

2.6.1 Utilization of waste WBP for the production of new PB

Czarnecki et al. (2003) investigated the possibility of substituting the particles in the core layer of PB with particles obtained from mechanically disintegrated waste WBP in a chipper. These waste WBP were classified as three-layer PB glued with urea formaldehyde (UF) or phenol formaldehyde (PF) binders and MDF glued with UF. The mechanically disintegrated particles/fibres were subjected to screening with 5.00 mm mesh size. These were blended with virgin particles in mass proportions of 10, 20, 30, 40, 50 and 60 % related to the total core particle mixture together with 8 % binder (UF or PF according to previously used binder type). Virgin pine particles obtained under industrial conditions were used in the surface layers together with 12 % binder. The produced PB had 700 kg m^{-3} density and 19 mm thickness. With respect to both adhesives, increasing the proportion of recovered particles obtained from UF-bonded waste PB in the core layer up to 50 % resulted in slight, yet systematic decrease of modulus of rupture (MOR). In contrast, even low proportion (10 % addition) of the recovered particles led to a remarkable decrease in internal bond strength (IB). Increasing amounts of the recovered particles resulted in a decrease in thickness swelling, and no significant changes in the free formaldehyde content (EN 120 1992). Particles obtained from mechanically disintegrated PF-bonded PB can even constitute up to 60 % the core layer without significantly worsening the examined properties. The best material properties, except MOR, were achieved when mechanically disintegrated particles from UF-bonded waste MDF were used together with virgin particles in the core layer of the PB.

Table 2.2: Published studies on the utilization of waste WBP with emphasis on producing PB, MDF and WPC.

Reference	Type of disintegrated waste WBP	Type of targeted end-product	Disintegration method
(Czarnecki et al. 2003)	PB, MDF	PB	Mechanical
(Martins et al. 2007)	WBP residuals	PB	Mechanical
(Himmel et al. 2014)	PB	PB	Mechanical
(Laskowska and Mamiński 2018)	PW	PB	Mechanical
(Azambuja et al. 2018)	PB, MDF, PW	PB	Mechanical
(Kraft 2007)	PB, FB	PB	Thermo-hydrolytic
(Demirkir and Çolakoğlu 2007)	PW	PB	Thermo-hydrolytic
(Lykidis and Grigoriou 2008)	PB	PB	Thermo-hydrolytic
(Lykidis and Grigoriou 2011)	PB	PB	Thermo-hydrolytic
(Roffael and Hüster 2012)	PB	PB-FB	Thermo-hydrolytic
(Ihnát et al. 2017)	PB, OSB	PB, MDF	Thermo-hydrolytic, mechanical
(Roffael et al. 2002)	PB, MDF	MDF	Thermo-hydro-mechanical
(Mantanis et al. 2004)	MDF	MDF	Thermo-hydrolytic
(Athanassiadou et al. 2005)	PB, MDF	MDF	Thermo-hydro-mechanical
(Roffael et al. 2010)	PB, FB	MDF	Thermo-hydrolytic
(Nicewicz and Danecki 2010)	FB	FB	Thermo-hydro-mechanical
(Klimczewski and Nicewicz 2013)	FB	HDF	Thermo-hydrolytic
(Roffael et al. 2016)	MDF	MDF	Thermo-hydrolytic
(Ihnát et al. 2018)	PB, MDF, OSB	PB, FB	Thermo-hydrolytic, Thermo-hydro-mechanical
(Roffael et al. 2018)	MDF	MDF	Thermo-hydrolytic
(Hong et al. 2020)	MDF	MDF	Thermo-hydrolytic
(Lubis et al. 2018b)	MDF	MDF	Thermo-hydro-mechanical
(Mitchell and Stevens 2009)	MDF	MDF	Thermo-hydro-electrical
(Moezzipour et al. 2018)	MDF	MDF	Thermo-hydro-electrical
(Dix et al. 2001a)	FB	MDF	Chemical-assisted
(Dix et al. 2001b)	PB, FB	MDF	Chemical-assisted-mechanical
(Schoo et al. 2003)	OSB	MDF	Chemical-assisted
(Balasuriya et al. 2003)	MDF dust	WPC	Mechanical
(Kamdem et al. 2004)	PB	WPC	Mechanical
(Chaharmahali et al. 2008)	PB dust, MDF dust	WPC	Mechanical
(Chavooshi and Madhoushi 2013)	MDF dust	WPC	Mechanical
(Madhoushi et al. 2013)	MDF dust	WPC	Mechanical
(Chavooshi et al. 2014)	MDF dust	WPC	Mechanical
(Migneault et al. 2014)	OSB dust, LVL dust	WPC	Mechanical
(Ashori et al. 2015)	MDF dust	WPC	Mechanical
(Gozdecki et al. 2015)	PB	WPC	Mechanical
(Krause et al. 2018)	PB, MDF	WPC	Mechanical
(Bütün et al. 2018)	MDF, HDF	WPC	Thermo-hydrolytic
(Bütün et al. 2019)	MDF	WPC	Thermo-hydro-mechanical, mechanical

Martins et al. (2007) studied the effect of utilising mechanically disintegrated waste WBP (including the production residues such as residual boards and board trimmings of WBP) on the formaldehyde content of PB. After mechanical disintegration in a chipper and raw material separation with respect to particle size distribution, new PB were produced with four different ratios of recovered particle content (30, 50, 70 and 100 %) together with virgin particles in the core layer with 7 % UF binder and solely virgin particles bonded with 7 % UF binder in the surface layers. All boards manufactured with higher percentages (≥50 %) of recovered particles displayed poor mechanical performance in comparison to boards with the standard mixture (30 % pine, 15 % eucalyptus, 25 % sawdust and 30 % recycled wood) and increased formaldehyde content, yet considerably below the maximum of E1 class according to (DIN EN 312 2003).

Himmel et al. (2014) studied the effects of mechanically produced recovered particles from waste PB on the formaldehyde release of new PB. To obtain homogeneous material, freshly produced PB (as simulated waste PB) were chipped to core layer particles in a hammer mill and served as core layer particles together with various proportions of virgin particles. Only (smaller) virgin particles were used for the surface layers. Nine different experimental PB were produced with pMDI to avoid the addition of formaldehyde via newly added UF adhesive. Thus, variants only differed in the core layer with respect to the proportion of recycled particles. The physico-mechanical properties of PB, which contained recovered particles in the core layer, did not differ significantly from those of boards made entirely from virgin wood particles, except for increased internal bond strength (IB) and decreased modulus of elasticity (MOE) both due to the decreasing aspect ratio. The authors attributed the differences in IB and MOE to the different particle morphology assuming that mechanically disintegrating the boards results in more spherical recovered particles than conventionally chipping the virgin wood particles. The total formaldehyde emission of the new PB positively correlated with the formaldehyde content of the recovered particles and could be predicted using regression models. Thus, due to the decreasing formaldehyde emission limits, the authors recommended to consider the contribution of both recovered and virgin wood particles as raw material with respect to the total formaldehyde release of the final PB. By using correlation analyses, it was possible to predict the formaldehyde emission of the new PB from the formaldehyde content of the individual raw materials when formaldehyde-free binder (pMDI) was used.

Another study examined the properties of PB produced from residual plywoods bonded either with UF or PF and compared them to PB made from virgin particles (Laskowska and Mamiński 2018). The panels were mechanically disintegrated using a wood shredder with 10.0, 14.0 mm (UF and PF), 25.0 and 38.0 mm (PF) mesh screens; thus, six grades of recovered particles were obtained depending on the mesh size and used binder type. These recovered particles were mixed to compose five different mixtures of recovered particles. Core layers of there-layer PB were prepared with five different ratios (20, 40, 60, 80 and 100 %) of these five recovered particles' mixtures and virgin particles. The type and content of the recovered particles strongly influence the resulting properties of the new PB. Although the results of the study were very divers, the authors concluded that UF and PF-bonded waste plywood can be recycled in the form of particles for new PB manufacturing using a shredder for mechanical disintegration.

Azambuja et al. (2018) mechanically disintegrated waste MDF, PB and plywood derived from construction and demolition waste using an industrial grinder and subsequently a hammer-mill with 8.0 – 14.0 mm mesh sizes. They used these particles for producing UF-bonded PB. New tree-layer PB containing a mixture of 25 % recovered particles obtained from each different waste WBP and 75 % virgin particles in the core layer displayed higher strengths and lower swelling in water than PB completely made from virgin particles.

Roffael and Roffael filed a patent (Roffael and Roffael 2004) on producing a binder based on extracted adhesive components from waste WBP blended with tannin components. The extracted adhesive components encompassed degradation products of UF resin such as urea, formaldehydes, methylolated urea and its oligomers as well as sugars extracted from the wood which are obtained from the thermo-hydrolytically disintegrated UF-bonded PB and/or FB in addition to polyphenolic ingredient extracted from spruce bark. Kraft (2007) intended for producing new PF-bonded PB, in which up to 20 % of the PF-binder was replaced by the aforementioned binder.

Demirkir and Çolakoğlu (2007) investigated the formaldehyde emission from PB manufactured with waste materials from the plywood industry. These used waste materials were peeling cores and dried veneer residues which were mechanically disintegrated by chipping, and residual plywood edge pieces containing UF resin. The latter were thermo-hydrolytically disintegrated in a digester at 110 °C for 20 min and chipped subsequently. Thereafter, new PB were produced using these recovered materials (solely or as a mixture of 33 % addition of each to virgin particles) bonded with UF binder and their formaldehyde content and emission were determined by the perforator method (EN 120 1992) and the flask method (DIN EN 717-3 1996). PB manufactured from 33 % containing thermo-hydrolytic disintegrated plywood edge pieces had 41 % lower formaldehyde content and 31 % lower formaldehyde emission value compared to PB manufactured from virgin particles. On the other hand, formaldehyde emission of PB manufactured from only mechanically disintegrated pieces and reference PB showed the highest formaldehyde values.

Lykidis and Grigoriou (2008) investigated the properties of PB made from recovered wood particles obtained by four different thermo-hydrolytic disintegration process conditions of PB. The processes varied with respect to applied pressure (0.2-0.8 MPa), temperature (119-167 °C) and duration (20-480 min) in an autoclave. The recovered particles contained a higher proportions of fine particles (< 1 mm), which is an indication that the disintegration process involves a reduction in particle size and can negatively affect some mechanical properties of the new PB. On average, PB produced from recovered wood particles showed statistically significant inferior mechanical and water-related properties compared to that of PB made from virgin particles with the exception of the MOE (in average 9.3 % increment) and formaldehyde content (in average 72.8 % reduction), especially when high temperatures and pressures were applied. Moreover, latter recycling processes were carried out under the same conditions as the first recycling processes for assessing the newly produced boards' properties. This process caused further quality degradation in the 2nd generation boards compared with boards produced after the first recycling processes, again with an exception of MOE. In addition, the recovered particles and, thus, the new PB obtained a darker colour due to thermo-hydrolytic disintegration. The authors recommended using mixtures of recovered and virgin wood particles for industrial application.

In another study (Lykidis and Grigoriou 2011), thermo-hydrolytic disintegration processes were carried out on laboratory scale in a closed vessel (with the capability to produce saturated steam, apply pressurized air and vacuum) at three temperatures (110, 130 and 150 °C), with three retention ratios of water (30, 45 and 65 % water uptake of the boards) as well as five process durations between 8 -75 min. The various recovered materials were used to produce PB with 12 mm thickness and 680 kg m^{-3} target density. Considering the physico-mechanical and chemical properties of the new PB, the optimum parameters for thermo-hydrolytic recovery were 45 % water retention at 150 °C temperature and 10 min treatment duration.

Roffael and Hüster (2012) studied the impact of thermo-hydrolytic disintegration of UF-bonded PB at two reaction times (3 and 24 h) applied at three temperatures (40, 103 and 150

°C) on the release of formaldehyde and volatile organic acids; formic acid and acetic acid assessed using the flask method (DIN EN 717-3 1996). Their results indicated that the formaldehyde emission of the recovered particles increases drastically with increasing disintegration temperature from 40°C to 150°C at 3 h reaction time. In addition, extending the reaction time from 3 to 24 h induced an extremely high increase in formaldehyde emission (91 and 81 % at 40 and 103 °C, respectively). The pH value of the absorption medium in the flasks decreased continuously when prolonging the reaction time at the same temperature or by increasing the temperature at the same reaction time due to production of volatile organic acids. Compared to the panels made of virgin particles, the release of formic acid was lower over the entire temperature range, while the emission of acetic acid soared to higher values.

Ihnát et al. (2017) described methods to recover particles from uncoated waste PB and waste OSB depending on their binder type (UF and MUF). They performed both dry mechanical disintegration using a drum chipper and thermo-hydrolytic disintegration of UF-bonded PB. For the latter process, they soaked the boards in cold water for 48 h first and then cooked them while stirring for 30, 60, 120 and 180 min to obtain recovered particles. HPLC analysis of the formaldehyde and urea contents in the wastewater extracts obtained after 30 min thermo-hydrolytic disintegration revealed 20 % hydrolysis of UF resin, which was removed from the particles. They also assumed a reduced amount of bonded formaldehyde in the recovered particles, because of the increased formaldehyde amount in the waste water extract. In order to disintegrate coated UF-bonded PB and to remove surface laminate, the panels were cooked in water for 120 min to additionally detach the coating. The obtained particles were further mechanically disintegrated using a drum chipper. Thermo-hydrolytic disintegration of MUF-bonded OSB was impossible even after 180 min of cooking. Therefore, these were mechanically disintegrated in a drum-mill chipper.

2.6.2 Utilization of waste WBP for the production of new MDF

Mantanis et al. (2004) conducted a study to partially replace virgin fibres with recovered fibres obtained from fibreboard production residues. They mechanically disintegrated residual boards of four different grades (standard MDF, moisture resistant MDF, colour impregnated MDF and hardboards) in a hammer mill and mixed the recovered fibres with virgin fibres in a 3:1 ratio. New MDF were produced with UF resin. Two compounds were added to the binder mix in three compositions, which the authors did not specify: a crosslinking agent (at levels 0, 5 and 10 % related to the total mass of the MDF) to enhance bonding and a key chemical agent (1 % related to the total mass of the MDF) for compatibility and efficient processing of virgin and recovered fibres. Increasing addition of the crosslinking agent improved the IB, MOR and thickness swelling, in average 28, 12 and 13 % respectively. The formaldehyde emission of all boards reached the requirements of the E2 class, because an E2 type UF binder was applied.

Roffael et al. (2002) and Athanassiadou et al. (2005) applied thermo-hydrolytic disintegration to UF-bonded waste PB and MDF under the high shear action of a twin screw extruder (TSE) containing 7 different compartments for obtaining suitable components for the production of new MDF. In the first part of the TSE, the solid waste material was fed and transported within the extruder through the so-called conveying screws. Thereafter, the conveyed waste material was subjected to thermo-hydrolytic disintegration at a temperature of 90 to 110 °C. One objective of this approach was to reactivate the UF resin used for bonding of waste panels. MDF containing recovered fibres with densities around 800 kg m^{-3} and 16 mm thickness displayed lower bending strength but comparable IB and thickness swelling as MDF respectively produced from virgin fibres. Using recovered fibres decreased formaldehyde emission of the boards, probably because the amino groups of remaining resin act as a formaldehyde scavenger. The formaldehyde emission of the new MDF made from both recovered and virgin fibres met the E1 class requirements.

Roffael et al. (2010) performed thermo-hydrolysis (at 170 °C for 5 min) of shredded UF-bonded PB and FB and subsequent mechanical disintegration in a refiner (TMP process) to produce new MDF. Morphological characterisation revealed shorter fibre length, lower cell wall thickness and higher content of fine fractions of the recovered fibres than of the virgin fibres. TMP fibres from waste panels exhibited higher pH value, contained higher amounts of acetate and formate in cold-water extracts and released higher formaldehyde emission assessed by the flask method compared to TMP fibres made from virgin wood. Still, the results indicated that up to 30 % of the virgin fibres in MDF could be replaced with the obtained recovered fibres without any noteworthy deterioration of the physico-mechanical properties. New MDF with 30 % recycled fibres even displayed a lower formaldehyde content determined by the perforator method (DIN EN 120 1992) than those made of 100 % virgin fibres.

Nicewicz and Danecki (2010) performed a thermo-hydrolytic disintegration process of waste adhesive-free insulation boards in a wet process. After an initial mechanical disintegration in a shredder, reduced pieces were then soaked in warm water and agitated for several minutes in a tank with an electric mixer and recovered fibres were obtained without further mechanical defibration. Recovered fibres and virgin fibres were mixed in 5 different ratios to produce new insulation boards in a wet process without binder and the formed mats were dried at 150 °C for 3 h. Increasing amounts of recovered fibre negatively affected the bending strength, while these reduced thickness swelling.

Klimczewski and Nicewicz (2013) assessed the fibre characteristics of different fibre materials. They mechanically disintegrated a mixture of waste fibreboards consisting of 80 % uncoated MDF, 10 % lacquered HDF and 10 % uncoated hardboards in a knife mill to pieces with less than 38 mm edge length and thermo-mechanically processed these in a refiner with the grinding gap set to 0.1 mm. Obtained recovered fibres were then used to produce HDF and were thereby compared to those used for standard HDF. The addition of recovered fibres caused a significant reduction in the fibre classes which was consisted of 90 classes depending on the fibre width (9 classes) and of fibre length (10 classes). The strength properties were negatively affected by a decrease in the proportion of fibre classes longer than 1711 µm and thickness swelling increased, when the classes of small fibres (200-956 µm) significantly increased.

Roffael et al. (2016) investigated the disintegration of mechanically shredded UF-bonded FB in a laboratory scale refiner (ca. 50 x 50 x 11 mm) at approximately 180 °C under 9 bar pressure for 4 min (TMP process). New UF-bonded MDF were produced by recovered fibres substituting 0, 33, 67 and 100 % by virgin fibres. Utilisation of 33 % recovered fibres for the MDF decreased IB by 26 % and reduced thickness swelling by 25 % compared to control MDF containing 100 % virgin fibres. Formaldehyde emission measured by the flask method was 56 % (3 h) and 46 % (24 h) lower for MDF containing 33 % recovered fibres than for the control boards. MDF containing 33 % recovered fibres displayed higher ammonia emission than respective control MDF (Roffael et al. 2018). MDF containing 67 and 100 % recovered fibres were produced only with hybrid bonding technology; combining 10 % UF-resin with either 0.5 or 1 % pMDI-resin. Supplanting virgin wood fibres with 67 and 100 % recovered fibres using hybrid bonding technology with 1 % pMDI lowered the IB (16 and 23 %, respectively), and formaldehyde content (72 and 84 %, respectively) while increasing the thickness swelling by 52 and 23 %, respectively. The authors concluded that thermo-hydrolytic disintegration liberates urea from the UF binder, which undergoes thermal decomposition in aqueous solutions and forms ammonium ions and ammonia gas. Thus, a significantly higher ammonia release was detected in the MDF containing 33 and 67 % recovered fibres to those containing 100 % virgin fibres.

Ihnát et al. (2018) cooked crushed waste UF-bonded MDF in water for 3 min with constant stirring and subsequently processed the obtained pulp in a refiner at a minimum temperature of 80 °C. In contrast, waste MUF-bonded MDF were first cooked in water for 180 min and then chipped in a drum chipper following by a refiner process at a minimum temperature of 80 °C. The fibre characterisation results indicated that the recovered fibres obtained from UF-bonded waste MDF and those from MUF-bonded waste MDF were suitable for the new MDF production. On the other hand, the formaldehyde content of the recovered fibres was reduced by 10-20 % (UF) and 30-40% (MUF) compared to the original amount of the waste MDF due to the hydrolysis.

Lubis et al. (2018b) studied the effect of mechanical disintegration after thermo-hydrolytic treatment of waste MDF on the quality of new MDF made thereof. Steam treated (at 180 °C under 6 bar pressure for 30 min) waste MDF pieces (30 x 30 x 25 mm) were either further processed in a refiner with a 1 mm plate gap for obtaining refiner recovered fibres (RRF) or in a hammer-mill for obtaining hammer mill recovered fibres (HRF). The average fibre length of both recovered fibre types was shorter than that of virgin fibres. Virgin fibres had higher coarseness compared to the recovered fibre types, while RRF was the finest. Both recovered fibre types had higher pH values than virgin fibres probably due to the remaining UF binder on the recovered fibres' surfaces and the formation of ammonia during its decomposition. HRF had a higher nitrogen content than RRF because of minor cleavage and solubilisation of the binder in the hammer-mill than in the refiner. Utilizing 100 % recovered fibres for the new MDF production resulted in lower density which might be due to the possible loss of fibres during mat forming, thus reducing the mechanical strength of the MDF. The results showed fluctuating trends for physico-mechanical properties of new MDF depending on the used recovered fibre. While MDF containing virgin fibres exhibited the lowest formaldehyde emission, MDF containing RRF showed much higher formaldehyde emission than those containing HRF.

Moezzipour et al. (2018) compared a thermo-hydrolytic (TH) disintegration process with a thermo-hydro-electrical (THE) process. After mechanical disintegration in a laboratory scale chipper, TH disintegration was applied in an autoclave at 105 °C for 150 min under 0.4 MPa pressure followed by defibrillation in a refiner. For the THE process, the waste MDF were chipped and the fragments were soaked in warm water for 30 min. After transferring the wet fragments into an open chamber, saltwater was added and electric current was passed through the mixture by electrodes. This led to the release of fibres. Recovered fibres obtained by the THE process had a similar length like virgin fibres but were longer than those obtained by the TH disintegration. Both types of recovered fibres exhibited significantly lower contents of extractives, hemicelluloses and lignin than virgin fibres, while the cellulose content was reportedly similar. Thickness swelling of MDF containing fibres recovered by the THE process was higher than that of MDF containing virgin fibres and lower than that of MDF containing fibres recovered by the TH process. Bending strength of MDF containing fibres recovered by the THE process was similar to those containing virgin fibres and higher than those containing fibres recovered by the TH process. IB of the MDF containing fibres recovered by the THE process was significantly higher than that of boards containing virgin fibres. MDF containing both types of recovered fibres emitted significantly less formaldehyde than those of virgin fibres, probably because UF binder residues acted as a scavenger.

Recently, Hong et al. (2020) investigated the effects of the addition of recovered fibres obtained by thermo-hydrolytic disintegration (30 kg of MDF pieces in 15 L water at 25 °C for 55 min with additional steam injection) of three different surface laminated MDF after an initial shredding in a hammer-mill and removal of any contaminants such as the surface lamination. These recovered fibres were added to the core layer (10, 20 and 30 % based on the core layer fibre mass) of the three-layer MDF and compared to MDF containing solely virgin fibres in the core layer. By increasing the content of the recovered fibres, regardless of the previous MDF-type, mechanical properties such as MOR and MOE and IB of the new MDF decreased and physical properties such as water uptake and thickness swelling increased. These results were attributed to the higher fine contents and higher pH of the recovered fibres compared to the virgin fibres.

Dix et al. (2001a) chopped laminate coated, UF-bonded waste FB in a chipper and subjected the panel fragments to a chemical-assisted thermo-hydro-chemical disintegration process (with 10 % NaOH solution at 200 °C for 2 h) comparable to alkaline pulping used for paper production (soda pulping). Newly produced MDF containing a mixture of 70 % virgin fibres and 30 % recovered fibres displayed higher IB and MOR than MDF solely produced from virgin fibres (control), while MDF containing 50 % virgin fibres and 50 % recovered fibres displayed similar mechanical properties as control MDF. The thickness swelling of MDF containing 30 and 50 % recovered fibres was even clearly lower than that of MDF derived solely from virgin fibres. As a follow-up study, Dix et al. (2001b) applied a comparable chemically assisted thermo-hydrolytic disintegration process (with 2 % Na_2SO_3 and 0.5 % NaOH at 180 °C for 5 min) comparable to CTMP pulping applied in the pulp and paper industry to obtain recovered fibres from waste PB and MDF. Newly produced MDF made from 100 % recovered fibres met the required standards (DIN EN 622-5 2009) with respect to the strength and swelling properties. While the strengths (IB, MOR) of MDF containing recovered fibres were up to 46 % lower compared to control MDF containing solely virgin fibres, the thickness swelling and the formaldehyde content of recovered MDF were respectively up to 55 and 73 % lower compared to control MDF.

Schoo et al. (2003) described a chemically assisted thermo-hydrolytic disintegration process of waste OSB, which were pMDI-bonded in the core and melamine-urea-phenol-formaldehyde (MUPF)-bonded in the surface layers. After mechanical disintegration of waste OSB, recovered fibres were obtained through a thermo-hydro-chemical process using a 10 % NaOH solution at 200 °C for 2 h. New MDF were produced, which contained 25 and 50% recovered fibres, and compared to respective panels solely made of virgin fibres (control). Using recovered fibres in both proportions reduced thickness swelling and increased IB of the MDF compared to control panels. Control and MDF containing 25 % of recovered fibres displayed similar MOR, while a proportion of 50 % recovered fibres reduced the bending strength by approximately 17 % compared to the control.

Mitchell and Stevens (2009) performed life cycle assessment to assess the environmental impacts of three waste MDF disposal routes based on thermo-hydro-electrical disintegration using microwave radiation. The three disposal routes involved landfilling, incineration without energy recovery and incineration with energy recovery. The study emphasises that the fibre production stage has the highest environmental impact when virgin fibres are used for MDF manufacturing. Thus, reducing the total inputs for virgin fibres by using recovered fibres should decrease environmental impacts. Moreover, when recovered fibres are used for MDF production, gas (fossil, natural and greenhouse) combustion would be significantly reduced or avoided, considering that much of the internally generated MDF waste is burned to produce process heat.

2.6.3 Utilization of waste WBP for production of new WPC

Balasuriya et al. (2003) studied the morphology and mechanical properties of compression-moulded WPC containing recovered particles (0, 20, 40, 50, 60 and 80 % related to the total mass of the composites) from waste MDF in dust form as fillers, virgin polyethylene (PE) as matrix and maleic anhydride modified polyethylene (MAPE) as coupling agent. Dust/particles derived from sawing, planing and sanding waste MDF. The morphology study of the produced WPC revealed that recovered particles dispersed well in the matrices. An increasing content of recovered particles (up to 50 %) induced an increasing crystallinity (up to 88.6 %) of the matrix polymer. This indicated that recovered particles were a potential nucleating agent for PE. FTIR spectroscopy, however, did not indicate chemical interactions between the recovered particles and PE. On the other hand, FTIR results reportedly indicated that addition of MAPE enhanced the compatibility of recovered particles and PE. The study shows that recovered particles obtained from waste MDF can replace virgin fibres as a potential reinforcing filler for polyethylene when MAPE is used as the matrix without significant property changes; utilization of recovered particles up to 50 % can be incorporated with clear improvement in mechanical properties of WPC.

Kamdem et al. (2004) produced compression-moulded WPC with virgin and recycled high-density polyethylene (HDPE) as matrix polymer and recovered particles from old CCA-treated red pine wood as well as waste UF-bonded PB. The two wood components were mechanically disintegrated in a hammermill; particles that pass through mesh size 30 (0.6 mm), but not through 100 (0.15 mm) were used. WPC containing recovered particles obtained from waste PB exhibited lower flexural bending strength and lower resistance to fungal degradation compared to WPC containing CCA-treated wood.

Chaharmahali et al. (2008) blended small particles (sawdust and grinding flour) from of PB and MDF with high-density polyethylene (HDPE) in a twin-screw extruder at 170 °C together with recycled HDPE in three different ratios (60, 70 and 80 % related to the total mass of the composites). MDF particles were much more fibrous than PB particles indicated by the longer dimension and higher aspect ratios of the former. Formulations containing recovered MDF particles exhibited slightly higher MOE than WPC containing recovered PB particles, which was attributed to the higher aspect ratios of the MDF particles. On the other hand, no significant difference in MOR was observed between WPC containing either one type of recovered particle or the combination of both at any given ratio. Screw withdrawal resistance values of formulations containing recycled PB were slightly higher than that of the other formulations at any given ratio. Meanwhile, all WPC panels had similar nail holding capacities. The Charpy impact strength was highest when recovered PB particles and recovered MDF particles were used together. All in all, very slight differences were observed between WPC containing only one type of recovered particles or the combination of both.

Chavooshi and Madhoushi (2013) studied the influence of various levels of MDF sawdust (40, 50, 60 % related to the total mass of the composites) and aluminium powder with polypropylene (PP) as matrix and maleic anhydride grafted polypropylene (MAPP) as coupling agent in various combination ratios on physico-mechanical properties of WPC produced by compression moulding. It was found that mechanical properties such as tensile strength, MOE and MOR decreased, while physical properties such as water uptake and thickness swelling increased with increasing amount of MDF dust and aluminium powder due to the poor compatibility between increasing wood and aluminium powder (up to 15 %) content with PP. A follow-up study investigated the effect of nano-clay particles in composites containing MDF dust (40, 50, 60 %), PP and 4 % MAPP produced by compression moulding (Chavooshi et al. 2014). Without nano-clay, increasing MDF dust content led to a reduction of screw and nail withdrawal strengths of the produced WPC and to a higher water absorption and thickness swelling. Addition of 2, 4 and 6 % (related to the mass of the composite) nano-clay, however, minimized the reduction of the screw and nail withdrawal strengths under wet conditions. Combined utilisation of nano-clay and the coupling agent MAPP as part of these WPC formulation had a positive synergetic effect on fungal decay resistance and reduced the water uptake compared to respective WPC without these additives (Ashori et al. 2015).

Madhoushi et al. (2013) studied the application of sanding dust from waste MDF (40, 50, 60 % related to the mass of the composites) as an alternative to virgin wood flour together with nano-clay for producing compression moulded WPC containing PP and 4 % MAPP. WPC containing higher MDF dust content (50 and 60 %) exhibited lower flexural and tensile strength compared to respective WPC containing 40 % MDF dust. The addition of 2 % nano-clay positively affected the mechanical properties of WPC irrespective of the MDF dust content, whereas addition of 4 and 6 % nano-clay decreased the mechanical properties due to the agglomeration of the clay particles in the composites. Meanwhile, physical properties such as water uptake and thickness swelling of the WPC improved by nano-clay addition in each formulation despite of increased MDF sawdust content.

Migneault et al. (2014) used sawing dust of OSB and LVL to produce injection moulded WPC with HDPE and 3 % MAPE. Adding dust from these panels provided roughly similar mechanical properties as those obtained with virgin wood dust, while WPC containing dust from the panel products showed faster water sorption than control WPC containing virgin wood dust.

Gozdecki et al. (2015) produced WPC from hammer-milled waste PB (20, 40 and 60 % related to total the mass of the composite) and polypropylene (PP) with and without adding 3 % MAPP as the coupling agent. The physico-mechanical properties of WPC containing particles from waste PB did not differ significantly from those containing virgin particles and fulfilled the standard physico-mechanical requirements for WPC.

WPC were produced by injection moulding with mechanically disintegrated UF-bonded PB and MDF particles in a hammer-mill (finally 1 mm mesh size; 60 % related to the total mass of the composite), PP (37 %) as matrix polymer and MAPP (3 %) as coupling agent (Krause et al. 2018). These particles exhibited higher aspect ratios than virgin wood particles and reprocessed WPC particles, which were obtained using the same consecutive hammer-milling process as for recovered WBP particles. As a result, WPC produced with the particles recovered from WBP showed improved mechanical properties than composites containing the two other particle types, especially when recovered MDF particles were used.

Bütün et al. (2018) performed thermo-hydrolytic disintegration of UF-bonded FB (MDF and HDF) at 95 °C in an autoclave and, after drying, subsequently shredded the obtained fibres in a hammer-mill (with 2.0 mm mesh size) to obtain small wood particles. WPC were produced by injection moulding containing this FB-derived particles or virgin wood particles (20, 60, 80 wt% related to total weight of composite) with polypropylene (PP) as matrix polymer with and without 3 % MAPP as coupling agent. Both types of WPC displayed similar physico-mechanical properties, but those of the composites containing recovered wood were somewhat better. With respect to mechanical properties, these differences were more prominent for bending properties and Charpy impact strength, while differences in tensile strengths were minor. These differences were attributed to enhanced compatibility between recovered wood particle surface and PP or to the higher flexibility of the recovered particles due to the influence of water at high temperature (95 °C) during the thermo-hydrolytic disintegration process.

In another study Bütün et al. (2019) compared the effect of recovered particles from MDF obtained by two disintegration methods: solely mechanical grinding using a hammer-mill with 2.0 mm mesh size and thermo-hydrolytic disintegration at 95 °C in an autoclave followed by grinding using a hammer-mill with 2.0 mm mesh size. The obtained particles were used to produce WPC by injection moulding with 60 % PP as well as with and without 3 % MAPP as coupling agent. WPC with both types of wood particles exhibited similar physico-mechanical properties but those obtained by thermo-hydrolysis imparted slightly enhanced strength and water-related properties. A reduced nitrogen content of the thermo-hydrolysed particles indicated a lower content of residual UF binder. A lower residual UF content might result in lower formaldehyde emission from WPC containing these particles during service life.

2.7 Conclusions

The main conclusions drawn from this review study are summarised as:

- Several processes are available: PB grinding is less costly and technically complex, while thermo-hydrolytic disintegration processes are better for FB to recover fibres. This is simply because grinding destroys the fibrous morphology, while the thermo-hydrolytic disintegration process promotes reactivation of the previously used resin.
- Waste WBP can principally be used to produce new WBP, in particular PB, MDF and WPC and partially and/or completely substitute virgin wood.
- However, recycling of particles and fibres may have a negative impact on mechanical properties of the newly produced MDF, because it affects the morphology of the wood material and UF binder residues might affect adhesion.
- UF binder residues may reduce the formaldehyde emissions.

- To produce PB and FB, recovered particles and fibres need to be combined with respective virgin material in order to minimise the reduction in the strength of the new PB and FB.
- However, the optimum proportion of recovered particles and fibres in PB and FB is not predictable.
- Particles obtained from waste WBP can be used to produce WPC with equal or even slightly enhance properties compared to WPC with respective virgin particles.
- Using sanding dust and sawdust of WBP in the manufacturing of WPC reduces production costs as well as eliminates the problem of accumulating and discarding wood industry waste.
- The utilization of recovered wood particles and fibres can maintain a favourable raw material supply for circular bioeconomy.

Chapter 3

Repeated thermo-hydrolytic disintegration of medium density fibreboards (MDF) for the production of new MDF

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Authorship

Table 3.0: Contribution of the co-authors (as modified from Hunt 1991).

	Planning & Design	Executing & Data Collection	Data analysis & Interpretation	Literary input	Σ
F. Yağmur Bütün Buschalsky	25	25	25	25	**100**
Carsten Mai	15	0	15	15	**45**

F. Yağmur Bütün Buschalsky prepared the experimental design together with Carsten Mai. She performed the data collection, analysis and interpretation. Furthermore, she wrote the manuscript.

Carsten Mai prepared the experimental design together with F. Yağmur Bütün Buschalsky. He assisted in data interpretation and manuscript writing.

3.1 Abstract

Medium density fibreboards (MDF) are currently not recycled after service life, but various publications report on recycling by the disintegration of MDF using various techniques and the properties of obtained recovered fibres (RF). In this study, the main aim was to put back RF into the MDF manufacturing process as closed-loop recycling using repeated thermo-hydrolytic disintegration. Compared to previous studies, we focus on the recycling of MDF with a relatively low F:U molar ratio (1.11). Urea-formaldehyde-bonded MDF with a target density of 700 kg m^{-3} were subjected to thermo-hydrolytic disintegration in an autoclave using only water at 95 °C for 20 to 30 min. Afterwards, the properties of RF and virgin fibres (VF), of MDF produced thereof and the composition of the disintegration water (DW) were determined. The nitrogen content (NC) revealed that RF contained about 30 % of the initially applied UF. The pH of the DW hardly changed during recycling and it contained considerable amounts of reducing sugars. Using RF did not result in higher formaldehyde emissions than VF. Compared to earlier studies using a higher formaldehyde content (higher F:U ratio), MDF bonded with modern UF resins can be disintegrated under clearly milder disintegration conditions with respect to temperature and time. The properties of recycled MDF were similar to those of reference MDF; up to 100 % RF could be used without severely deteriorating the strength and increasing formaldehyde emissions from these panels.

3.2 Keywords

Fibreboard; MDF; recycling; recovered fibres; mechanical properties; physical properties; formaldehyde emission

3.3 Introduction

Medium density fibreboard (MDF) is a dry formed engineered wood product composed of fine lignocellulosic fibres, combined with a synthetic resin and joined under heat and pressure (Thoemen et al. 2010). MDF is used in a wide range of applications mainly for flooring, but also for construction and furniture. It has substituted solid timber as well as other wood-based panels (WBP), due to its availability and wide range of thicknesses and the ability to be machined and finished to a high standard (WPIF 2014). Thus, fibreboard production in Europe encompasses mainly MDF, which makes up about 70 % of the fibreboard market in Europe (Mantanis et al. 2018). Global MDF production, on the other hand, together with high density fibreboard (HDF) has reached 98.6 million m^3 in 2018 (FAO 2019a). After service life, these panels will eventually become waste that is deposited in landfills or burned (Irle et al. 2018). Incineration of waste MDF for energy production, in the form of heat rather than electricity, is used as a waste disposal option (Mitchell and Stevens 2009). Even though the word “waste” projects a perception of a material with no value or useful purpose (Rowell et al. 1991), these panels and production residues can be disintegrated using various techniques and the obtained recovered fibres (RF) can be utilized as an alternative wood source for further material production such as MDF.

Hitherto, three types of techniques have been developed for disintegrating thermosetting resin-containing WBP. These include mechanical, thermo-hydrolytic and chemical-assisted disintegration techniques. The thermo-hydrolytic disintegration technique is viable for WBP that are bonded with hydrolysable resins (such as urea-formaldehyde - UF). After crushing the material, liquid water and/or steam at high temperature and pressure causes hydrolytic cleavage of resin bonds in WBP (Kharazipour and Kües 2007). Thus, the process detaches the wood particles (or fibres) constituting roughly 90 % of the WBP, which can thus be further processed.

Thermo-hydrolytic disintegration of MDF may alter the properties of the wood fibres, which are designated as recovered fibres (RF) after this process. Changed fibre morphology (e.g. aspect ratio), chemical nature of the fibres and residual resin might influence panel properties such as strength, water uptake and thickness swelling (Benthien et al. 2017). Therefore, utilising RF for new MDF production can affect the physico-mechanical and chemical properties of the panels. It was recently shown that 10 % recovered fibres obtained from offcuts and trims of high density fibreboard (HDF) can be added to virgin fibres to produce industrial HDF, which meets the related standard requirements (Sala et al. 2020). On the other hand, the disintegration water (DW), which is removed after the thermo-hydrolytic disintegration process, contains a certain amount of hydrolysed UF resin, and most likely some degraded wood components. Only two previous studies (Hagel and Saake 2020, Lubis et al. 2018a) related to thermo-hydrolytic disintegration of WBP have investigated the DW. Knowing the chemical composition of the DW is especially important to balance the process streams and to pursue wastewater management.

Previous studies about waste WBP disintegration have dealt with panels containing UF resins with a higher formaldehyde content (higher formaldehyde to urea molar ratio) than nowadays. However, in the late 80s, resin manufacturers turned towards decreasing the F:U molar ratio in order to reduce formaldehyde emission (Markessini 1994). Thus, the results of more previous studies are based upon data between 2001 and 2010. In addition, most studies in this field have only focused on a single disintegration cycle. This study assesses the recyclability of MDF with lower F:U molar ratio. RF obtained from thermo-hydrolytic disintegration of uncoated MDF were put back into the MDF manufacturing process as a closed loop recycling. The main aim was to investigate the effects of using RF on the properties of newly produced MDF after repeated thermo-hydrolytic disintegration processes. Furthermore, the study examines the chemical properties (nitrogen, formaldehyde, pH) of the RF compared to virgin fibres (VF) and relates it to the properties of the respective DW.

3.4 Material and Methods

3.4.1 Production of the first-generation MDF panels

Virgin fibres (VF) of 90 % Scots pine (*Pinus sylvestris*) and 10 % radiata pine (*Pinus radiata*) were obtained from Steico SE (Feldkirchen, Germany). Prior to resinating, fibres were loosened in a hammermill (VS1 N, Electra SAS, Poudenas, France) using 2.0 mm mesh size. A proportion of 10 % (dry mass of resin) commercial liquid urea-formaldehyde (UF) resin (Kaurit 350, BASF, Ludwigshafen, Germany) based on dry fibre mass and 2 % ammonium sulphate ($(NH_4)_2SO_4$) based on the resin solid content as hardener were sprayed onto the fibres in a drum blender with rotating speed at 50 rounds min^{-1}. After resinating, the fibres were once again loosened in the aforementioned hammermill with the same mesh size. Subsequently, fibre mats were manually formed using a 450 x 450 mm^2 forming box on an aluminium plate covered with siliconized paper and pre-pressed. A siliconized paper and a second aluminium plate were laid on the pre-pressed mat for pressing the panels under heat (190 °C) and pressure (5 N mm^{-2}) to a target density of 700 kg m^{-3} to reach a panel thickness of either 10 mm or 16 mm using metal bar stops. The pressing time amounted to 15 s mm^{-1}. A total of 15 MDF panels were produced for each targeted thickness (Table 3.1). After a day of cooling down and subsequent trimming the edges of the panels to 400 x 400 mm^2, the panels were cut to dimensions depending on further processing and stored in a climate chamber at 20 °C with 65 % relative humidity until they reached a constant mass.

Table 3.1: Production of the MDF panels throughout the different generations.

MDF generation and thickness	Used fibre type	Number of produced MDF	Number of produced reference-MDF using virgin fibres
1st generation, 10 mm	Virgin fibres	15	-
1st generation, 16 mm	Virgin fibres	15	-
2nd generation, 10 mm	RF from thermo-hydrolytic disintegration of 10 mm 1st generation MDF	8	2
2nd generation, 16 mm	RF from thermo-hydrolytic disintegration of 16 mm 1st generation MDF	9	3
3rd generation, 10 mm	RF from thermo-hydrolytic disintegration of 10 mm 2nd generation MDF	5	3
3rd generation, 16 mm	RF from thermo-hydrolytic disintegration of 16 mm 2nd generation MDF	6	3

3.4.2 Thermo-hydrolytic disintegration of MDF panels

For each disintegration run, pieces cut from first or subsequent generation MDF with 50 x 50 mm^2 in cross-section were immersed in water at 95 °C in the autoclave Zirbus Z3 (Zirbus Technology GmbH, Bad Grund, Germany) for a certain duration depending on the board thickness (Table 3.2). After removing the excess water (so-called disintegration water - DW) using a centrifuge device (a spin-dryer for laundry 776 SEK, Robert-Thomas GmbH, Neunkirchen, Germany) recovered fibres (RF1, RF2, the number denotes the recycling runs) were dried to 5 % moisture content at 70 °C in a drying-oven. From this, it can be reasonably assumed that each kilo of RF material absorbed ca. 2 litres of the disintegration water.

Table 3.2: Thermo-hydrolytic disintegration process parameters of the MDF panels throughout the different generations.

Thermo-hydrolytic disintegration run	Disintegrated MDF panel	Amount of disintegrated MDF (kg)	Amount of water at 95 °C (l)	Duration (min)
A	1st generation, 10 mm	4	28	20
B	1st generation, 10 mm	4	28	20
C	1st generation, 10 mm	4	28	20
D	1st generation, 10mm	4	28	20
E	1st generation, 10 mm	4	28	20
F	1st generation, 16 mm	5	32	30
G	1st generation, 16 mm	5	32	30
H	1st generation, 16 mm	5	32	30
I	1st generation, 16 mm	5	32	30
J	1st generation, 16 mm	5	32	30
K	1st generation, 16 mm	5	32	30
L	2nd generation, 10 mm	4	28	20
M	2nd generation, 10 mm	4	28	20
N	2nd generation, 10 mm	1.5	10.5	20
O	2nd generation, 16 mm	5	32	30
P	2nd generation, 16 mm	5	32	30
R	2nd generation, 16 mm	5	32	30
S	2nd generation, 16 mm	3	18	30

3.4.3 Production of the second and third generation MDF panels

Second and third-generation MDF were produced using RF from thermo-hydrolytic disintegration of the previous generation MDF. The production process, methods, materials and their amounts were kept identical to the previous MDF generation. Second-generation MDF of 10 mm thickness were produced using RF1 obtained from thermo-hydrolytic disintegration of 10 mm first-generation MDF – equivalently for 16 mm second-generation MDF and the following recycling run (Table 3.1). A total of 8 (10 mm thickness) and 9 (16 mm thickness) MDF panels were produced. In parallel, 2 (10 mm thickness) and 3 (16 mm thickness) reference-MDF panels were produced from virgin fibres. The total numbers of third-generation MDF panels using RF2 were 5 (10 mm thickness) and 6 (16 mm thickness). In addition to the recycled third-generation MDF, 3 reference-MDF panels were produced for each targeted thickness using virgin fibres.

3.4.4 Chemical properties of the VF and RF

The pH value was measured after cold water extraction of the fibres using two replicas; 2 g of sample material was placed into a 100 ml Erlenmeyer flask and 60 ml of distilled water was added. Afterwards, the Erlenmeyer flask was placed on a shaker at room temperature, the measurements were done after 20 min, 4 h and 24 h with a pH electrode InoLab Level 2 (WTW GmbH, Weilheim, Germany). Formaldehyde emission from the fibre mass was determined based on the basic principle of the standard EN 717-3 (1996) using approximately 2 g of fibre material in a bleached tea bag after 3 h and 24 h emission. NC was determined using the Kjeldahl method as described previously (Bütün et al. 2019) in order to assess the remaining resin content on the RF after thermo-hydrolytic disintegration. Calculation of UF content assumed that the UF resin contains roughly 30 % of nitrogen. Morphological characterization of the RF and comparison with virgin fibres were separately studied (F.Y.B.B., unpublished data 2020).

3.4.5 Determination of the physico-mechanical and chemical properties of the MDF panels

The density of each individually cut MDF specimen was determined according to EN 323 (1993) and the overall density of a panel was calculated as the average of these samples. Moisture content (MC) was determined according to EN 322 (1993) with 12 specimens per panel. Internal bond strength (IB) was determined according to EN 319 (1993) and thickness swelling (TS) and water uptake (WU) after 24 h immersion in water according to EN 317 (1993) applying sample dimensions of 50 x 50 x board thickness (mm^3) with 3 specimens per panel. Flexural strength and modulus of elasticity (MOE) were determined according to EN 310 (1993) applying sample dimension of 250 x 50 x 10 mm^3 and 370 x 50 x 16 mm^3 depending on the board thickness with 3 specimens per panel. Formaldehyde emission of the boards was analysed according to EN 120 (1992, perforator method) and EN 717–2 (1995, gas analysis method) with a single specimen per panel and EN 717–3 (1996, flask method) with two specimens per panel for 3 h and 24 h. Nitrogen content (NC) of the panels was determined using Kjeldahl method as described previously (Bütün et al. 2019). The UF content of the produced MDF panels was calculated considering that the UF resin contains roughly 30 % of nitrogen neglecting the very low NC of the virgin fibres (< 0.1%).

3.4.6 Chemical analysis of the DW

Disintegration water (DW) was analysed with respect to the pH value, formaldehyde content based on acetylacetone method (EN 120 1992), NC based on Kjeldahl method as described previously (Bütün et al. 2019) and the UF content was calculated (considering that the UF resin contains roughly 30 % of nitrogen) for samples after each disintegration run. The amount of reducing sugar equivalents in the DW was assessed based on 3,5-dinitrosalicylic acid (DNSA) method (Miller 1959).

3.4.7 Statistical analysis

Welch's t-test was conducted at a level of significance $\alpha = 0.05$ to determine the significance of the results among the panels throughout the different generations using Excel 2016 (Microsoft, Redmond, WA, USA). Except for the NC and calculated UF content of the MDF panels and RF, each thickness class was evaluated individually. Second- and third-generation MDF panels were compared to their corresponding first-generation MDF panels and to their reference panels depending on the panel thicknesses.

3.5 Results and Discussion

3.5.1 Chemical properties of the virgin (VF) and recycled fibres (RF)

The pH values of the fibres were measured after three different time spans but the differences for each fibre type were so low that only the average value was reported (Table 3.3). While the pH value of VF amounted to 4.0, those of RF were in average 4.9 and 5.2 after each recycling process, respectively. The slightly higher pH values of the RF may be due to the formation of ammonia due to the breakdown of the UF-resin, which reacts to alkaline ammonium hydroxide in water (Roffael and Hüster 2012; Wan et al. 2014). Roffael et al. (2009) assumed, that the highly increased pH value of the RF (6.3) in their study also enhances the cleavage of acetyl groups in hemicelluloses and subsequently of the glycosidic bonds.

Formaldehyde emission of the VF after 3 h was 1.8 and 2.0 mg per kg oven-dried fibres for non-milled and hammer-milled samples, respectively, without significant difference (Table 3.3). Each recycling process increased the formaldehyde emissions of the RF. In addition, the hammer-milling process further increased the emission, except after hammer-milling of second-time recycled RF of 16 mm panels. The 24 h formaldehyde emission of non-milled and hammer-milled samples VF were again the same resulting in 8.6 and 10.0 mg per kg oven-dried fibres, respectively.

Virgin fibres' NC of 0.07 % is in accordance with Cowling and Merrill (1966), who reported that wood contains approximately 0.1 % nitrogen; Lubis et al. (2018a) found 0.5 % nitrogen in VF. While the NC of the VF was neglicibly low, the NC of the RF amounted to approximately 1 % after the first and 2 % after the second disintegration processes (Table 3.3). As the NC of the virgin MDF panels was approximately 3 % (Table 3.4), this indicates that about 33.5 % of the UF binder remained on the first-generation RF and 53.4 % on the second- generation RF (related to the virgin panels).

3.5.2 Physico-mechanical and chemical properties of the MDF panels

MDF produced from RF exhibited slightly higher densities than the target density and respective reference panels (Table 3.5). Even though, the IB of the second-generation panels was identical to that of the first-generation and corresponding reference panels. Moreover, the IB of the third-generation 10 mm MDF panels showed 64.5 % increment (at slight density increase of 3.8 %), while 16 mm MDF panels showed 89.5 % increment (the density increased by 4.0 %) compared to corresponding first-generation panels. In addition to the slightly increased density that may affect IB (Wong et al. 2000; Hong et al. 2017, 2020), the main reason for the higher IB might be the increasing amount of adhesive that remains on the fibres after recycling as previously reported (Roffael et al. 2002, 2010; Lykidis and Grigoriou 2008; Bütün et al. 2018; Lubis et al. 2018b). On the other hand, Roffael et al. (2016) reported that the substitution of VF by 100 % RF decreased the IB of the new MDF significantly. This might

Table 3.3: Chemical properties of the virgin fibres (VF) and recovered fibres (RF). Equal letters within a property indicate that there is no statistical difference between the fibre types.

Fibre type			pH (20 min, 4 and 24 h)	Formaldehyde emission- Flask method (mg/kg o.d. fibre) (3 h)	Formaldehyde emission- Flask method (mg/kg o.d. fibre) (24 h)	Nitrogen content (%)	Calculated UF content (%)	Amount of initial UF content remained on the fibres (%)
Virgin fibres	Non-milled		4.0^{a}	1.8^{a}	8.6^{a}	0.07^{a}	0.2^{a}	n.a.
	Milled		4.0^{a}	2.0^{a}	10.0^{a}	0.07^{a}	0.2^{a}	n.a.
RF 1	10mm	Non-milled	4.8^{c}	50.5^{b}	595.0^{b}	1.1^{b}	3.5^{b}	35.0
		Milled	4.8^{bc}	61.3^{b}	557.0^{b}	1.0^{b}	3.2^{b}	32.0
	16mm	Non-milled	4.9	51.0^{c}	465^{c}	1.2^{b}	3.8^{b}	38.0
		Milled	5.2^{d}	62.9^{c}	517.0^{c}	1.1^{b}	3.5^{b}	35.0
RF 2	10mm	Non-milled	5.3^{b}	61.2^{b}	617^{b}	2.2^{c}	7.0^{c}	51.8
		Milled	5.0	79.5^{b}	629.0^{b}	2.5^{c}	8.0^{c}	59.2
	16mm	Non-milled	5.4^{d}	75.5^{c}	617.0^{c}	2.2^{c}	7.0^{c}	54.6
		Milled	5.4^{d}	68.0^{c}	561.0^{c}	1.9^{c}	6.1^{c}	47.6

n.a.= “not applicable”

be due to strength loss of the recycled fibres and the over proportional consumption of adhesive by fines, which occurs more in recycled MDF. Moreover, a slight reduction of fibre length, which may occur during recycling as reported elsewhere (F.Y.B.B., unpublished data 2020), might explain higher IB. Shorter fibres may result in a larger surface area for enhanced adhesion. In addition, short fibres can strengthen the structure of the MDF by filling the voids between the large fibres, while aligning themselves along the forming direction (Back 1987). Correspondingly, Groom et al. (2002) suggested that there is an optimal ratio of fines (shorter fibres) to larger fibres, where the fines provide a greater degree of bonding. Shortening of fibres, however, reduces flexural strength (Myers 1983; Ye et al. 2007) but a reduction in MOR and MOE was hardly observed (Table 3.5). On the other hand, flexural strength (MOR) and MOE of recovered MDF panels (10 mm thickness) were not significantly different despite increasing density (on average 3 % related to first-generation panels) throughout the different generations. Furthermore, repeated recycling of 16 mm MDF panels resulted in rather fluctuating results in terms of bending strengths, which were not significant and were attributed to fluctuations in the production process. In terms of MOE, 16 mm MDF showed similar trends except for a 5.8 % decrement for the third-generation panels in comparison to the corresponding reference panels instead of an increment.

Thickness swelling (TS) of the MDF panels after 24 h immersion in water progressed differently to the water uptake (WU) (Table 3.5). The second-generation 10 mm panels showed 16 % and 10 % reduced TS compared to the corresponding reference and first-generation panels, respectively. TS of third-generation 10 mm panels was same as the corresponding reference and first-generation panels. Recycled 16 mm MDF, on the other hand, showed constantly higher TS throughout the generations in comparison to the corresponding reference and first-generation panels. The WU of the MDF after 24 h immersion did not exhibit significant differences between 10 mm second- and third-generation panels and their corresponding reference panels, but significantly higher values when compared to the first-generation panels (Table 3.5). The WU of 16 mm panels were identical between second- and third-generation panels. The recycled 16 mm panels showed higher water uptake than the first-generation and the reference panels. Higher WU and TS values of the recycled MDF panels are usually attributed to the poor adhesion of the panels, which can be detected by lower IB. In this study, even though the IB increased throughout the repeated recycling processes, the WU and TS increased after recycling. Moezzipour et al. (2018) assumed the removal of the lignin as a reason; however, this is unlikely for the present study due to neutral pH value and a relatively moderate temperature during disintegration. A possible reason for higher WU and TS might be the increased availability of hygroscopic low-molecular-weight sugars and of fines, which might act like wicks. In contrast to the present findings, Roffael et al. (2016) observed reduced WU and TS when VF were substituted with 33 % RF. They attributed this improvement to the reactivation of the sizing agent (water repellent) contained in the recycled industrial waste MDF and cross-linking of the RF by free formaldehyde in the UF resin. Virgin panels in the present study, however, did not contain sizing agents. In addition, the higher amount of adhesive in the recovered MDF and the resulting increase in IB might explain the stable water-related properties between the 2nd and the 3rd generations of the recycled panels.

Urea, ammonia and oligomeric decay products of UF resin can react with formaldehyde and act as formaldehyde scavengers. Thus, various studies reported significantly reduced formaldehyde content and emission of the recycled MDF (Roffael et al. 2010, 2016; Behn et al. 2018). Similarly, the formaldehyde emission determined by perforator method of the recycled panels was lower than that of virgin panels throughout the different generations of this study. However, formaldehyde emission results assessed by the gas analysis and flask method did not follow the same trend.

The formaldehyde emission determined by perforator method of the MDF revealed different trends depending on the panel thicknesses (Table 3.4). Still, regarding both thicknesses, the first-generation panels displayed a significantly higher formaldehyde emission compared to the later generations and their reference panels and the subsequent recycling further decreased the formaldehyde content of the recycled panels. Recycled 10 mm panels had an identical formaldehyde emission to their reference panels. However, 16 mm recycled panels exhibited significantly lower formaldehyde content of 15.4 % (second-generation) and 18.8 % third-generation) compared to their corresponding reference panels. Likewise, formaldehyde emission assessed by gas analyser showed different trends depending on the panel thicknesses. Again, the formaldehyde emission of the first-generation panels was significantly higher than that of the subsequent generations and their reference panels.

Table 3.4: Chemical properties of virgin and recycled MDF panels. Asterisks and equal letters within a property indicate that there is no statistical difference between the MDF panels throughout the different generations.

MDF panels			Formaldehyde content- Perforator method (mg/100g o.d. board)	Formaldehyde emission- Gas analyser method (mg/m^2h)	Formaldehyde emission- Flask method (mg/kg o.d. board) (3h)	Formaldehyde emission- Flask method (mg/kg o.d. board) (24h)	Nitrogen content (%)	Calculated UF content (%)
1st generation	10 mm		17.2	14.8^{a}	12.2^{a}	162^{a}	3.1^{a}	10.0^{a}
	16 mm		17.7	14.5^{d}	12.0^{c}	163^{d}	3.1^{a}	10.0^{a}
2nd generation	10 mm	Recycled	15.2^{ad}	10.9	9.3^{b}	209^{b}	4.2	13.5
		Reference	15.1^{ace}	12.3^{c}	10.0^{b}	182^{abc}	3.2^{a}	10.4^{a}
	16 mm	Recycled	13.7^{f}	11.2^{e}	8.8^{d}	114^{e}	4.0	12.8
		Reference	16.2	10.9^{e}	9.6^{de}	135^{ef}	3.0^{a}	9.9^{a}
3rd generation	10 mm	Recycled	13.4^{be}	12.9^{bc}	9.6^{b}	148^{c}	5.4	17.3
		Reference	14^{bcd}	13.6^{abc}	13.6^{ab}	173^{a}	3.2^{a}	10.3^{a}
	16mm	Recycled	11.6	13.9^{d}	10.3^{e}	136^{df}	4.9	15.7
		Reference	14.3^{f}	13.4^{de}	12.7^{c}	180^{def}	3.1^{a}	9.9^{a}

Formaldehyde emissions of both thickness-type panels were lower after the subsequent recycling processes, while that of 10 mm panels was always lower in comparison to their corresponding reference panels. Panels of 16 mm thickness showed the opposite tendency with slightly but not significantly higher formaldehyde emission values compared to their reference panels.

Formaldehyde emission after 3 h assessed by the flask method also revealed similar trends to the ones observed by perforator and gas analysis methods; first-generation panels released significantly higher emissions than the subsequent generations and their reference panels. Furthermore, the 10 mm recycled and respective reference panels displayed identical formaldehyde emission after 3 h. Similarly, 16 mm recycled panels and second-generation reference panels were identical in terms of 3 h formaldehyde emission. Reduction of formaldehyde emissions due to recycling is in contrast to increasingly higher emissions of the RF. This might be explained by the fact that the decay products of UF resin (urea, ammonia and oligomers) in the RF material are able to react with formaldehyde and act as formaldehyde scavengers.

Interestingly, the formaldehyde emissions during 24 h measured by flask method resulted in a different tendency to other related methods. When compared to the first-generation panels, 10 mm and 16 mm recycled panels showed unequal trends; emission of 10 mm second-generation panels was 22.5 % (significantly) higher compared to the first-generation panels, while that of third-generation panels was 8.6 % lower. The formaldehyde emission of second-generation 16 mm panels, however, was 30.1 % and that of third-generation panels 16.5 % lower than the first-generation panels. Both in second and third generations, however, 16 mm recycled panels were not significantly different from their reference panels. Compared to the previous studies (Dix et al. 2001a, b; Schoo et al. 2003; Roffael et al. 2010), the rather milder conditions (lower temperature and/or shorter duration) might result in a lower breakdown of the UF resin during the thermo-hydrolytic disintegration. Thus, the slight increment in the formaldehyde emission of the third-generation panels can be due to the higher amount of remained non-hydrolysed UF-binder, even though, these values were always lower than the first-generation panels, except the 24 h flask method results for 10 mm second-generation MDF panels.

The nitrogen content (NC) of the first-generation and reference MDF panels amounted to 3.1 % and indicate that the targeted 10% UF resin content was reached. NC was determined as an indicator for the UF content of the recycled panels. It exhibited a constant increment after the subsequent recycling processes, while these values were the same for the first-generation and reference panels irrespective of the panel thicknesses. As a result of the remaining UF binder on the RF (33.5 % in RF1 and 53.4 % in RF2), the NC increased by 35.4 % and 74.2 % related to virgin panels for 10 mm recycled panels and by 29.0 % and 58.0 % for 16 mm recycled panels after each disintegration process, respectively.

A possible explanation for the inconsistent physicomechanical and fluctuation of chemical results between 10 and 16 mm panels may be attributed to the natural variability of the wood quality along with the limitations of the laboratory-scale production of them.

3.5.3 Chemical characteristics of the disintegration water (DW)

The pH values of the DW were increased from 4.5 to 5.1 over the various disintegration runs (Table 3.6). The formaldehyde content in the DW after the first disintegration process was 249.5 mg l^{-1} and 342.3 mg l^{-1} after recycling of the 10 mm and 16 mm first-generation panels, respectively. An increment of 11.0 % was observed after the subsequent recycling of the 10 mm second-generation MDF panels, while 33.4 % decrement occurred after the recycling of the 16 mm second-generation panels.

Significant levels of reducing sugar equivalents (6-7 mg l^{-1}) were detected in the DW after the first disintegration process. These might originate mainly from hemicelluloses, which were cleaved during the production (pulping) of VF or from panel recycling. During these processes cleavage of acetyl groups may occur followed by cleavage of glycosidic bonds mainly of hemicelluloses. After the second disintegration process, however, the amounts of reducing sugar equivalents in DW were lower than 1 mg l^{-1} showing 91.4 % and 87.0 % decrement for 10 mm and 16 mm panels, respectively (Table 3.6). This indicates that the first disintegration process already removed most of the extractable sugars. The reason, why the pH values of the DW obtained after the first disintegration process, is acidic, is that the VF exhibit a pH value of 4.0 (Table 3.3) due to formation of acetic acid during pulping. During the second disintegration process, the pH values of DW was higher, but still below that of demineralised water.

Table 3.5: Physico-mechanical properties of virgin and recycled MDF panels. Asterisks and equal letters within a property indicate that there is no statistical difference between the MDF panels throughout the different generations.

MDF panels			Density (kg m^{-3})	Internal bond strength (IB) (N mm^{-2})	Flexural strength (N mm^{-2})	Modulus of elasticity (MOE) (N mm^{-2})	Water uptake (WU) (%) 24h	Thickness welling (TS) (%) 24h
1st generation	10 mm		727^{a}	0.31^{a}	30.4*	2992*	68.6^{a}	19^{a}
	16 mm		742^{c}	0.19^{c}	30.6^{a}	3310^{a}	62.6^{c}	17.3^{d}
2nd generation	10 mm	Recycled	743^{b}	0.34^{ab}	29.6*	3020*	77.8^{b}	17.1
		Reference	739^{ab}	0.22	32.2*	3447*	76.4^{ab}	20.4^{abc}
	16 mm	Recycled	766^{d}	0.23^{d}	32.3^{ac}	3536^{c}	71.2^{d}	18.6^{eg}
		Reference	773^{d}	0.19^{cd}	34.9^{c}	4019^{d}	53.6^{ce}	16.2^{de}
3rd generation	10 mm	Recycled	756^{b}	0.51	28.5*	2750*	73.7^{b}	19.8^{c}
		Reference	717^{b}	0.39^{b}	28.6*	2974*	75.6^{ab}	21.2^{b}
	16 mm	Recycled	773^{d}	0.36	35.2^{bc}	3451^{abcd}	71.5^{d}	19.5^{f}
		Reference	747^{c}	0.20^{cd}	34^{bc}	3667^{bc}	54.2^{e}	16.8^{dfg}

The NC of the DW obtained after the disintegration of the second-generation 10 mm panels was slightly higher (7.0 %) than after the previous disintegration. Meanwhile, 16 mm panels had a slightly lower (6.4 %) NC in DW obtained after the second disintegration. Based on the assumption that, each kilo of RF material absorbs ca. 2 litres of water, in average, approximately 65 % and 50 % of the initial UF was found in the DW samples from the 1st and the 2nd disintegration processes, respectively. The NC of a specific recycled MDF should be roughly the sum of the NC of the used RF and ca. 3.1 % nitrogen, which derives from the 10 % targeted UF resin. The difference between the total NC of the recycled boards and the NC of DW and RF combined is attributed to evaporation (Hagel and Saake 2020) during draining of the DW and/or during the RF drying.

Table 3.6: Chemical properties of the disintegration water (DW) obtained after the disintegration processes compared to the demineralised water.

Chemical properties	Disintegration water				Demineralised water
	1st disintegration process		2nd disintegration process		
	Runs for 1st generation, 10 mm MDF	Runs for 1st generation, 16 mm MDF	Runs for 2nd generation, 10 mm MDF	Runs for 2nd generation, 16 mm MDF	
	A, B, C, D, E	F, G, H, I, J, K	L, M, N	O, P, R, S	
pH	4.5	4.7	5.0	5.1	5.2
Formaldehyde content (mg l^{-1})	249.5	342.3	277.0	227.8	n. a.
Reducing sugars and equivalents (mg l^{-1})	7.0	6.2	0.6	0.8	0.2
Nitrogen content (%)	0.4	0.47	0.43	0.44	<0.003
Calculated UF content (%)	1.2	1.5	1.4	1.4	n. a.

n.a.= “not analysed”

3.6 Conclusions

The aim of the present study was to assess the recyclability of MDF bonded with current standard UF resins with a relatively low F:U molar ratio using repeated thermo-hydrolytic disintegration. The process required to disintegrate the panels used significantly lower temperatures and was clearly shorter than in previous studies. Even the utilisation of 100 % RF did not bring about significantly lower strength or higher formaldehyde emissions than that of virgin boards. Utilising up to 100 % RF may be an alternative environmentally friendly source for new MDF production. Repeated recycling processes may enhance IB and reduce formaldehyde emissions from the new panels. The thermo-hydrolytic disintegration process alters the chemical properties of the fibres resulting in higher pH and causes formaldehyde emission from the RF due to the remaining UF-resin. The initially high amount of reducing sugar equivalents in the DW after the first recycling indicates the cleavage of the hemicelluloses during fibre production (pulping) and/or thermo-hydrolytic recycling.

Conflict of interest

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Chapter 4

Geometric changes of TMP fibres due to thermo-hydrolytic disintegration of waste medium density fibreboards (MDF) evaluated with three fibre analysers

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Authorship

Table 4.0: Contribution of co-authors (as modified from Hunt 1991).

	Planning & Design	Executing & Data Collection	Data analysis & Interpretation	Literary input	Σ
F. Yağmur Bütün Buschalsky	25	20	20	25	**90**
Arne Imken	0	5	5	0	**10**
Stergios Adamopoulos	0	0	10	10	**25**
Sheikh Ali Ahmed	0	5	0	0	**5**
Burkhard Plinke	0	0	15	15	**30**
Carsten Mai	10	0	10	10	**30**

F. Yağmur Bütün Buschalsky prepared the experimental design together with Carsten Mai. She executed the data collection together with Arne Imken and Sheikh Ali Ahmed. Furthermore, she performed the data analysis and interpretation with Stergios Adomopoulos, Burkhard Plinke and Carsten Mai, and she wrote the manuscript.

Arne Imken executed the data collection together with F. Yağmur Bütün Buschalsky and Sheikh Ali Ahmed.

Stergios Adamopoulos supported data analysis and interpretation and assisted manuscript writing.

Sheikh Ali Ahmed executed the data collection together with F. Yağmur Bütün Buschalsky and Arne Imken.

Burkhard Plinke supported data analysis and interpretation and assisted manuscript writing.

Carsten Mai prepared the experimental design together with F. Yağmur Bütün Buschalsky. He assisted in data interpretation and manuscript writing.

4.1 Abstract

Thermo-hydrolytic disintegration of medium density fibreboards (MDF) is a promising method for recovering fibres that can be used, for example, for the production of new MDF. This disintegration process, however, may affect the geometry and size distribution of the applied fibres initially obtained by thermomechanical pulping (TMP) and, thus, the properties of MDF newly produced thereof. Optical methods based on image acquisition are increasingly common for geometrical characterisation to assess the quality of fibres. In this study, we analysed the size distribution of recovered fibres (RF) obtained by thermo-hydrolytic disintegration of urea-formaldehyde bonded waste MDF and subsequent hammer-milling by using three optical fibre analysers, namely FibreShape Pro (I.S.T.), QICPIC (Sympatec) and Valmet FS5, while comparing them to virgin fibres (VF) obtained by TMP. The three analysers use different evaluation methods and therefore, as expected, provided different absolute values with regard to the fibre geometries. Therefore, size distributions could not be directly compared. Nevertheless, the results provided an indication of the changes that occurred during the manufacturing and recycling process. RF displayed a very similar fibre geometry compared to the originally used VF, in spite of process-related shortening. Average fibre length (geodetic length, GL) and length-based distribution of VF were always greater than those of RF, while fibre width (geodetic width, GW) of the hammer-milled RF was slightly greater than that of VF. The three analysers, however, provided substantially different results regarding average fibre length and fibre length distribution, especially between FibreShape Pro and QICPIC, although the general tendencies observed for the process related changes were consistent.

4.2 Keywords

MDF; recycling; fibre length; morphology; particle analyser; recycling

4.3 Introduction

Medium density fibreboard (MDF) is an engineered wood panel product with a worldwide production of more than 100 million m^3 per annum (FAO 2019a, 2020), which is made by hot pressing of thermomechanical pulping (TMP) virgin wood fibres produced in a refiner and mostly urea-formaldehyde (UF) resin as a binder. Today, the commercially viable processes to recycle MDF or furniture made from MDF are very limited, while it is done industrially for particleboards through a simple grinding process. In order to preserve the fibre shape and dimension, the adhesive bonds between fibres need to be removed by hydrolytic processes (Irle et al. 2018). Previous studies to recycle MDF involved hydrolysis of UF by applying steam and high temperature (Riddiough 2002; Sandison 2002; Michanickl and Boehme 2003; Kearley and Goroyias 2004; Nakos et al. 2005; Lykidis and Grigoriou 2008; Beele 2009; Mitchell and Stevens 2009; New 2012; Wan et al. 2014; Moezzipour et al. 2018). Shortcomings had to do with the complexity of methods, high energy consumption, batch processes and high labour costs, low removal levels of the cured resin, and low quality of the RF (Roffael et al. 2010; Lubis et al. 2018a).

The morphology and size distribution of wood particles and fibres are important for the production and the resulting properties of wood-based panels (Benthien et al. 2014b; Ohlmeyer et al. 2015). The dimensions of wood fibres may be steered in the production process, e.g. by adjusting the gap between the refiner discs and their plate design. Fibre material used to produce MDF consists of shorter and thinner fibres than that used for insulation panels of much lower density (Imken et al. 2021). With MDF, it is mainly the fibre length that determines the modulus of rupture (MOR) of the boards (Steffen et al. 2001; Dix et al. 2001a; Benthien et al. 2014a). In this respect, the recycling process may affect the physical (such as fibre length), mechanical (such as flexibility) and chemical properties (such as nitrogen content, pH etc.) of fibres (Rowell et al. 1993). Previous approaches of MDF recycling have not yet fully explored the geometrical changes of the RF material obtained after waste MDF recycling.

Typical fibres for application in composites have high strength and stiffness in fibre direction allowing high load transfer, while they are weak and flexible if load is applied perpendicular to it (Steinmann and Saelhoff 2016). When studying mechanical properties of the MDF, the area-weighted distribution of the fibres is more relevant than the other related distributions, because strength and Young’s modulus modelling are based on fibre and (spherical) particle volume fractions (Le Moigne et al. 2011). However, the assumption of spherical shapes does not fit for wood fibres due to their large aspect ratios (width to length ratio). Wood fibres tend to be curved, and therefore it is challenging to determine their actual length and width. TMP fibres tend to agglomerate in clumps (formation of felted fibre balls) during sieve analysis (Plinke et al. 2012), which is thus not applicable (Plinke et al. 2016). Moreover, sieve analysis reflects the probability of particles passing through a sieve mesh, and this probability is determined by the width of individual particles rather than their length.

Automatic measurement of single fibres in digital images is technically possible if the fibres are distributed without overlap and their shapes can be recognized with sufficient accuracy (Plinke et al. 2016). Digital image analysis (DIA) is often used as the sole method to determine the size and shape of fibres or to verify and complement results obtained from other methods (Teuber et al. 2016b). In fact, the DIA method provides far more data values within an acceptable time than any of the other conventional methods such as manual fibre inspection or sieve-based methods (e.g. air-jet sieve) but it is a totally different approach to fibre size evaluation and therefore the characteristics determined are hardly comparable, at least for fibres. Mechanical size measurement devices like sieves supply one distribution representing the probability for each particle to pass the sieve. This makes only sense for more or less “compact” particles, but MDF fibres do not behave like wood particles from a hammer mill or a flaker – they agglomerate and have to be dispersed before the measurement. An image analysis system assesses the shape of the fibre based on the longest and shortest diameters, perimeter, and projected area or by equivalent spherical diameter. These methods for characterizing the size distribution of fibres have only recently become available as commercially applicable systems (Plinke et al. 2016). All of these systems take contour/binary images of the fibres and analyse them using standard image processing techniques to obtain size and shape parameters of each fibre and provide data for statistical evaluation. Nonetheless, important sub-procedures may differ between various analysis systems with respect to the methods for fibre dispersion, pre-processing of the images, size parameters determination and processing speed (Plinke et al. 2016). Fibre dispersion can proceed in a dry form or as an aqueous dispersion. Dry dispersion is favourable for free-flowing fibres with the help of a vibrating active (compressed air) dispersion unit (Teuber 2016), wet dispersion is more suitable for finer fibres, such as in the paper industry, where fibres are measured in a suspension to ensure a proper material separation. Dispersion by compressed air or water is much more effective than passive or manual dispersion.

Optical methods based on image acquisition developed to determine particle and fibre dimensions provide only geometric and morphological features of the fibres “as seen by the camera” but not mechanical properties like e.g. strength and stiffness, and no features beyond the optical resolution of the sensor, such as surface roughness. According to ISO 13322 Part 1 (ISO 13322-1 2014) and Part 2 (ISO 13322-2 2014) image analysis methods may be divided into static and dynamic methods. As a static method, FibreShape Pro (Innovative Sintering Technologies Ltd., Vilters, Switzerland) is a scanner-based system, which is cost-efficient because it does not require an expensive image capturing system (Gusovius et al. 2006). It is described to be easy to apply and reliable for measuring fibre length, width, and fineness, and other fibre and fibre product qualities (Müssig and Schmid 2004). Dynamic methods obtain the images of the samples from moving objects (Teuber 2016). Devices such as QICPIC (Sympatec, GmbH, Clausthal-Zellerfeld, Germany) try to achieve the fibre separation through a capillary tube applying airflow in which the individual fibres are illuminated by a light source and on the opposite side, a highly sensitive camera is located for recording the images. The primary results from image analysis are based on number distributions. The number-weighted distribution represents the number of fibres that make up a given fraction of fibre with respect to the length or width or aspect ratio; it weights each counted particle or fibre equally. The area-weighted distribution corresponds to the projection area taken by fibres that make up a given fraction of the fibre length and aspect ratio (Le Moigne et al. 2011); thus, bigger fibres account for a higher proportion of the material. Considering that the fibre length correlates with eminent parameters such as refiner and bulk density, using the length-weighted distribution is recommended for reducing the influence of the fines content within the sample (Hirn and Bauer 2006). Absolute comparability between different optical methods and devices cannot be guaranteed, as these methods differ in terms of dispersion and in the algorithms for determining the size parameters (Plinke et al. 2016).

Up to now, several studies have been published on the comparison of fibre length analysers (Guay et al. 2005; Hirn and Bauer 2006; Cöpür and Makkonen 2007; Plinke et al. 2016), from various fields of application, especially for papermaking and production of wood polymer composites (WPC). This study analysed the influence of the thermo-hydrolytic disintegration of waste MDF on the geometry and length distribution of recovered fibres by using three types of fibre length analysers; FibreShape Pro – a scanner-based system, QICPIC – a measuring system for dry fibres using image acquisition in a measurement chamber, and Valmet FS5 - using image acquisition directly in the sample suspension with a “diving” camera.

The results of the analysis are compared to those of the virgin fibre material initially applied to produce the MDF. Changes in RF geometry were expected as a result of forces applied during thermo-hydrolytic disintegration process. The general characteristics of the used analysers are listed in Table 4.1.

4.4 Material and methods

4.4.1 Attainment of the recovered fibres

First-generation MDF were produced with virgin wood fibres (90 % Scots pine, *Pinus sylvestris* L., and 10 % radiata pine, *Pinus radiata* D. Don) with 10 % UF resin (Kaurit 350, BASF, Ludwigshafen, Germany) based on dry fibre mass and 2% ammonium sulphate ($(NH_4)_2SO_4$) as hardener based on the resin solid content. The resinated fibre material was hot-pressed (190 °C, 5 N mm^{-2}, 15 s mm^{-1}) to obtain panels of 10 mm and 16 mm thickness bearing a target density of 700 kg m^{-3}. The first type of recovered fibres (RF1-10 and RF1-16) were obtained by thermo-hydrolytic disintegration of these MDF. The disintegration processes were carried out in an autoclave in which each 1 kg of MDF pieces (50 x 50 mm^2 cross-section) were heated in 7 L water at 95 °C for 20 min (10 mm MDF) and in 6.4 L water at 95 °C for 30 min (16 mm MDF). After drying of the RF to 5 % moisture content at 70 °C, these were loosened up in the hammer-mill (VS1 N Electra SAS, Poudenas, France) using 2.00 mesh size. Second-generation MDF containing the first type of hammer-milled RF were produced as above. Second-generation MDF of 10 mm thickness were produced using hammer-milled RF1-10 and second-generation MDF of 16 mm thickness using hammer-milled RF1-16. Same as above, thermo-hydrolytic disintegration of the second-generation MDF provided second type RF (RF2-10 and RF2-16), which were again dried and loosen up by hammer-milling, thus, third-generation MDF containing RF2 were produced accordingly. Samples from all RF types and VF were taken before and after hammer-milling for geometrical analysis (Figure 4.1). Images of all samples were taken using a digital light microscope (VHX 5000, Keyence Deutschland GmbH, Neu-Isenburg, Germany) (Figure 4.2).

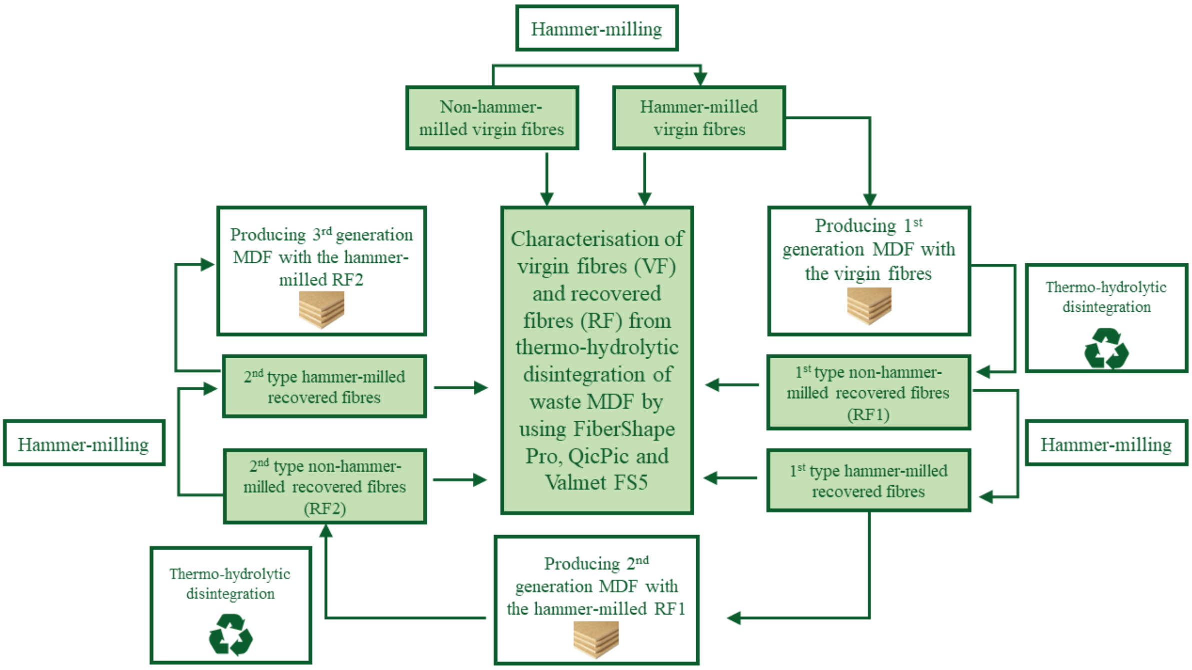

Figure 4.1: Schematic diagram of the produced MDF generations, their thermo-mechanical disintegration and obtained recovered fibre types. Ligth green highligted boxes envision the geometrical characterisation of the fibre material (non-hammer-milled and hammer-milled virgin fibres and recovered fibres).

All fibres including VF were conditioned at 20 °C and 65 % relative humidity and subsequently characterised using three devices (Table 4.1).

Table 4.1: Characteristics of the chosen fibre analysers.

Model and manufacturer	Dispersion	Range (µm)	Optical resolution	Principle	Image acquisition method
FibreShape Pro, Innovative Sintering Technologies	Dry dispersion, manual separation	30 – 300 to 40 – 10.000	100 – 3.200 dpi (6.400 dpi) 21 µm/pixel	Fibre length determination based on rectangle model	Static
QICPIC, Sympatec	Dry and wet dispersion, automatic fibre separation via air jet sieve	0.55 – 33,792	1024 x 1024 pixels of 10 µm x 10 µm area with 256 grey levels (8 bit)	Fibre length measurement by skeletonizing	Dynamic
Valmet FS5, Valmet	Wet dispersion	10 – 7,600	1296 – 966 pixels with 256 grey levels	Fibre length measurement along the fibre centreline, classification into different fractions and fines	Dynamic

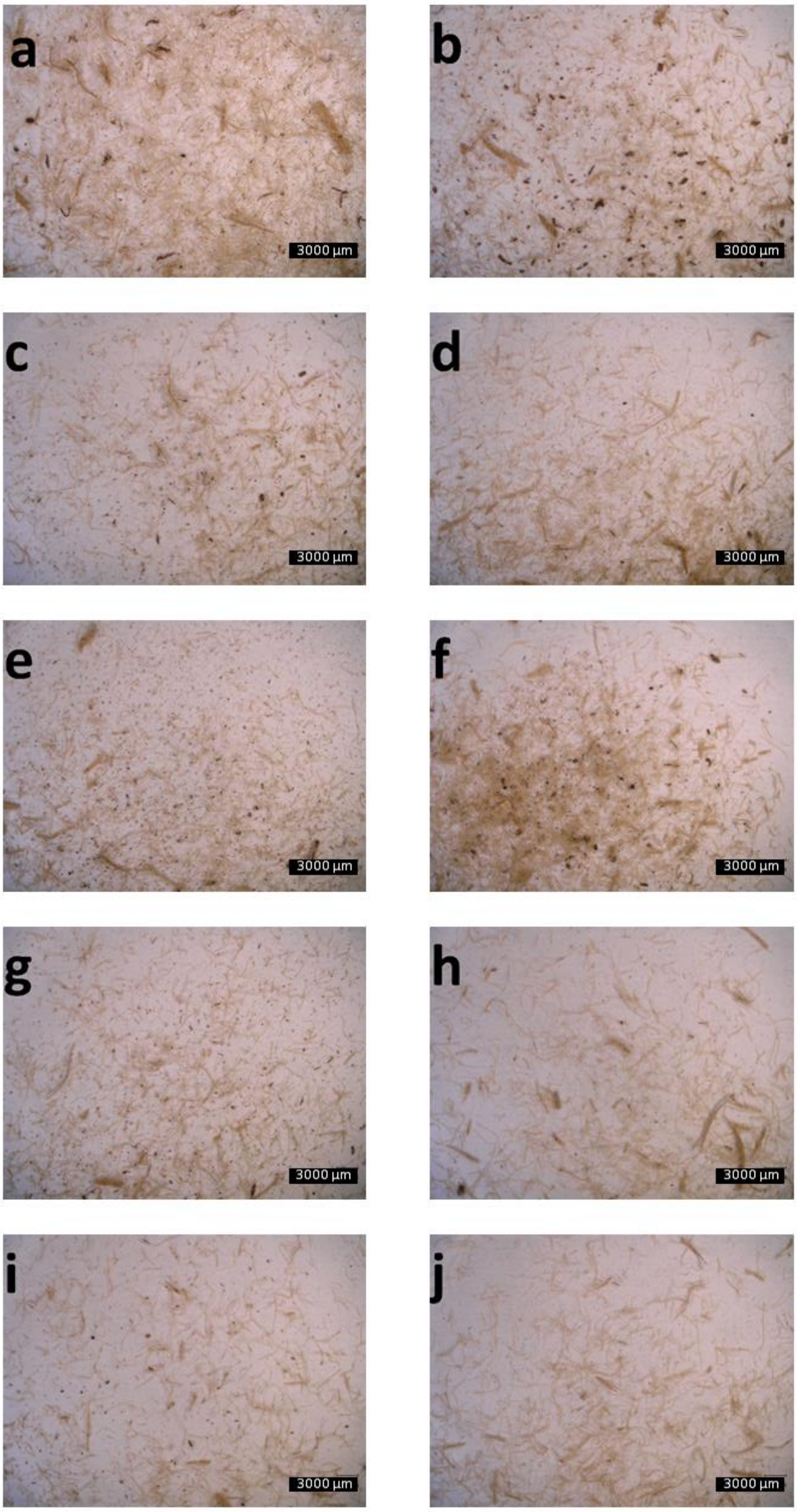

Figure 4.2: Images of analysed fibres a) virgin fibres non -milled, b) virgin fibres, milled, c) RF1-10, non-milled, d) RF1-10, milled, e) RF2-10, non-milled, f) RF2-10, milled, g) RF1-16, non-milled, h) RF1-16, milled, i) RF2-16, non-milled, j) RF2-16, milled.

The fibre analysers QICPIC and FS5 do not allow to measure a sample more than once – after the measurement the smaple has to be discarded. However, each sample comprises several thousands of fibres, therefore the statistics are reliable with regard to the individual sample, but during sampling it has to be made sure that the sample is representing the material.

4.4.2 Scanner based image analysis of fibres using FibreShape Pro

Two replicas of 0.25 g from each fibre sample (each before and after the hammer-milling) were evenly dispersed on a flatbed scanner glass using a strainer; overlapping fibres were separated using tweezers. V850 Pro scanner (Epson AG, Suwa, Nagano, Japan) and the scanner software SilverFast Ai Studio (LaserSoft Imaging AG, Kiel, Germany) were used to acquire images of the samples. The scanned images were analysed using FibreShape Pro (quality control, research and development version 6.1.4) software (Innovative Sintering Technologies Ltd., Vilters, Switzerland). The FibreShape Pro software detects the individual fibres and measures their contour area and perimeter of the fibres' shapes based on the ISO (International Organisation for Standardisation) standards (ISO 9276-1 2004; ISO 9276-6 2008). A standard 10-10,000 µm measuring mask was used with 1200 dots per inch (dpi) image resolution that corresponds to a pixel size of approx. 21 µm. The contour area was given by the number of pixels belonging to a closed shape, the contour length was computed by following the shape and counting the outer pixels. The FibreShape software determines the length and width of each particle by applying the "rectangle model" (Schmid and Schmid 2006) to these values: length and width of each fibre were computed from the values of its area and contour length, using the formulas for area and contour length of a rectangle as functions of its length and width. By experience, this is a good compromise for the determination of length and width even for particles with a complex shape such as fibres – although many other definitions for "length" and "width" are possible. The particle size distribution was characterized using the median (x_{50}) and two other most common distribution indices on the x-axis (percentiles x_{10} and x_{90}) of the length-based particle size distribution (Q1). The quantification of the length proportion of fibres that fall under a percentile was enabled. Length-weighted particle size distribution was chosen in order to make the results comparable to the other methods. It was calculated automatically from the histogram data by the software, where the added length of the particles in each size class was measured and its percentage of the total added length of all particles in the sample calculated

(ISO 9276-1 2004). In addition, the analysis provided the geodesic fibre length (GL), which is the length-weighted average fibre length. The area ratio (area of all objects/total image area) was kept under 3.5 %, following the manufacturer’s recommendation to reduce the amount of overlapping fibres. Two replicate measurements were recorded for each fibre type. Examined fibres were recollected attentively and kept in containers for further analysis with QICPIC.

4.4.3 Dynamic image analysis of fibres using QICPIC

Fibre samples collected after FibreShape Pro measurement were analysed in the dynamic image analysis system QICPIC (Sympatec, GmbH, Clausthal-Zellerfeld, Germany) combined with the dry dispersion unit RODOS (Sympatec, GmbH, Clausthal-Zellerfeld, Germany). Due to the non-free-flowing nature of the fibres, they were first manually dispersed evenly on a tray and sucked into the device with a flexible hose attached to the dry dispersion unit. Fibres were then dispersed in an accelerated laminar air-jet and pictures were taken from the fibre stream with a high-speed CMOS (complementary metal-oxide-semiconductor) camera. The pressure of the air-jet breaks up agglomerations and ensures that the fibres enter the measurement zone in a random orientation; this reduces the number of overlapping fibres (Witt et al. 2004). The software WINDOX (Sympatec, GmbH, Clausthal-Zellerfeld, Germany) is used to measure the fibre length from the projected fibre area of binary pictures and to calculate the size distributions (Teuber et al. 2016). The WINDOX software considers the shortest path between the most distant endpoints as the length of the fibre and “skeletonizes” this path into sections, while considering the projected area divided by the sum of all segments as the width of the fibre (Witt et al. 2007). This is a different approach compared to the rectangle model used by FibreShape. As for FibreShape Pro, particle length was characterized in terms of the most common distribution width on the x-axis (percentile x_{10}, median (x_{50}) and x_{90}) of the length-based particle size distribution (Q1).

4.4.4 Analysis of fibres in aqueous suspension using Valmet FS5

Fibre analyses in aqueous suspension were performed by means of the Valmet FS5 fibre image analyser (Valmet Automation Oy, Espoo, Finland). About 0.2 g of each fibre type was suspended in 5l distilled water. In the device, an aliquot of 500 g of this suspension was pumped through a capillary tube (0.2-0.4 mm in diameter) in which a high-definition camera takes a certain number of images (typically 2000) (in grayscale). From these images, parameters such as length and width distribution are determined by a computer algorithm (Padberg et al. 2016; Taylor et al. 2020). Geodesic fibre length (GL, measured by following the centre line of each fibre) and average fibre width (GW) were used for the comparison. The device is designed to analyse fibres in papermaking processes and therefore also assesses the data on the number of counted fibres, cell wall thickness, and the number of fibre kinks, kink angles, gravimetric coarseness, fibre curl and the proportion of fines but these are not shown. The fibre types RF1-10, non-milled and RF2-16, non-milled were not analysed due to the lack of sampling amount. Two replicate measurements were recorded for virgin fibres, while RF types were measured only once.

4.4.5 Statistical analysis of fibre size and shape distributions

The length-weighted distributions, aspect ratio and GL have been statistically analysed using MS Excel 2016 (Microsoft Corporation, Redmond, Washington, USA). One-way (single factor) analysis of variance (ANOVA) was conducted at a level of significance $\alpha = 0.05$ to determine the significance of a possible effect of thermo-hydrolytic disintegration process on the fibre geometry by using the different devices.

4.5 Results and discussion

4.5.1 Length distribution, average length / width and aspect ratio

Manual dispersion of the fibres for analyses with FibreShape Pro required more preparation and thus, took much longer time than for the other two devices as reported previously (Plinke et al. 2016; Benthien et al. 2017). The three analytical devices provided some different results (Table 4.2). The comparison of the same fibre samples revealed major differences between the analysers, especially between FibreShape Pro and QICPIC. Although the fibre material tends to tangle and its agglomerates could not be actively dispersed on the scanner of FibreShape Pro (Plinke et al. 2016), the analyser did not provide greater length values than Valmet FS5. On the contrary, FibreShape Pro generated lower values at the absolute level. The difference between absolute average fibre lengths (GL) determined by FibreShape Pro and Valmet FS5 was approximately 9.7 %, while Valmet FS5 mostly provided higher values. A relative comparison of GL determined by these two analysers exhibited a high consistency; this fact was approved by the statistical analysis. FibreShape Pro indicated 86, 77, and 75 % higher values regarding x_{10}, x_{50} and x_{90}, respectively, and 20 % lower aspect ratio. These differences might be attributed to the different dispersion methods (manually in FibreShape, by air or water in QICPIC and FS5) image acquisition (scanner vs high-speed camera) and length determination methods of the devices. In addition, different optical resolution between the analysers might result in the different ability of fine particles' recognition. This might be the main reason for the contrasting length-based distribution values between FibreShape Pro and QICPIC analysers. Another possible explanation is that with the same fibre type, deviations in measured values between the analysers are inevitable, while with the same analyser between different fibre types deviations are reliably different (Plinke et al. 2016). By experience there is no "ground truth" in particle measurement since each device and method uses its own algorithms to determine a few indices to characterize a complex reality. Therefore, equal results between devices cannot be expected, but trends between samples and experiments should be reflected. Nevertheless, the individual devices are suitable for monitoring (relative) changes during the recycling process, even if the absolute fibre lengths cannot be determined with certainty.

4.5.2 Effect of thermo-hydrolytic disintegration

Average fibre length and length-based distribution of virgin fibres measured by all analysers were always larger than the RF, while fibre widths of hammer-milled RF (except RF2-10, milled) were slightly wider (ca. 4.6 %) than those of hammer-milled virgin fibres (Table 4.2). On the other hand, some hammer-milled samples (RF1-16, milled and RF2-16, milled) gave higher GL values (both generated by FibreShape Pro and Valmet FS5) than the respective non-milled samples (RF1-16, non-milled and RF2-16, non-milled). This might be due to an increase in the number of small fibres below the detection limit after mechanical disintegration process. Thus, these small fibres were neglected in the weight factor of the longer fibres increases when calculating the average fibre length (GL) and results in a higher GL value.

In other words, when the full range of the fibre length was considered (including the fine fractions), RF samples contained much higher percentages (i.e. numbers) in the smaller fractions (Figure 4.3 and Figure 4.4). Analysis with Fibreshape Pro revealed that about 50 % of non-milled and milled virgin fibres exhibited a length in the range of 1700 – 9500 µm and 1500 – 7300 µm, respectively. However, only ca. 30 % of the fibres in the RF were in these aforementioned ranges. This indicates a deterioration of the fibre geometry, in particular a shortening of the fibres, due to the severe thermo-hydrolytic disintegration conditions. Although a box plot representation of the average fibre length shows very similar distributions for all fibre types (Figure 4.5), the cumulative frequency (Figure 4.3) and the percentage representation of the fibre length (Figure 4.4) indicated narrower distributions for the virgin fibres and broader distributions for the RF.

With respect to the influence of refiner fibre quality on the properties of injection moulded wood-plastic composites, Schirp et al. (2014) suggested that, in addition to fibre length and aspect ratio, other factors have to be taken into consideration, such as total fibre surface area and topography as well as the chemical composition of the fibre surface. This is especially important when considering that RF usually still contain UF binder due to uncompleted hydrolysis and subsequent removal, which may affect the morphology of RF. Different results (GL and GW) obtained for different fibre types (VF vs. RF) consolidates this importance. Especially the wider GW of RF might be explained by the remaining binder, which is discussed in one of the published study of the co-authors (Bütün Buschalsky and Mai 2021)

In addition, fibre bundles could still be found in RF but hammer-milling resulted in a complete fibre separation. Obtained RF maintained a fibre morphology (the distinctive difference between length and width directions) rather than a spherical particle morphology. Nevertheless, analysis with FibreShape Pro revealed that RF still tended to entangle and agglomerate in fibre balls, akin to virgin fibres.

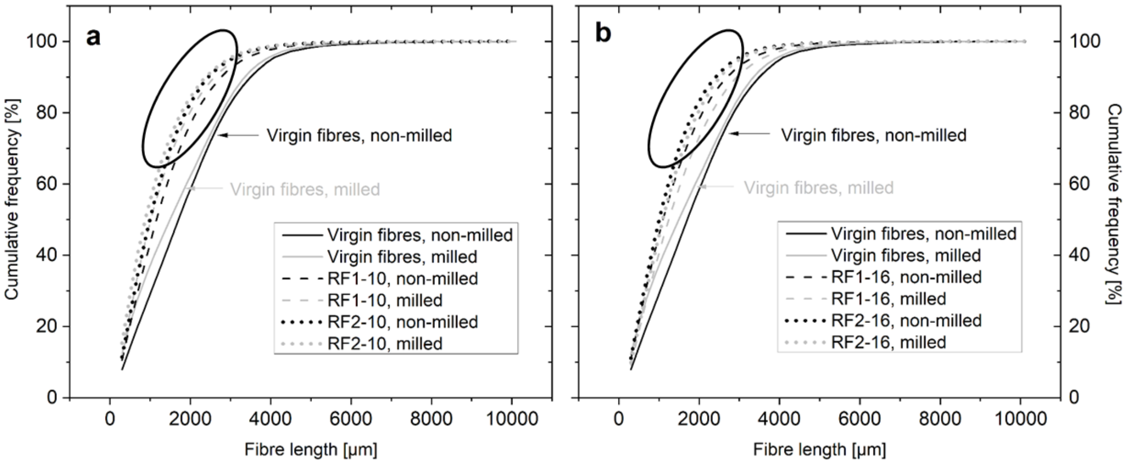

Figure 4.3: Cumulative frequency of the fibre length distribution based on FibreShape Pro for a) virgin fibres vs. recovered fibres obtained from disintegrating 10 mm MDF and b) virgin fibres vs. recovered fibres obtained from disintegrating 16 mm MDF. The oval highlighted areas show that the recovered fibres contain much higher percentages (thus, numbers) in the smaller fractions.

Table 4.2: Summary of the geometrical analysis based on the fibre length-based distribution (Q1) concerning the median (x_{50}), the 10th and the 90th percentiles (x_{10} and x_{90}) as well as the aspect ratio (length/width) for FibreShape Pro and QICPIC, geodesic fibre length (GL) for FibreShape Pro and Valmet FS5 and fibre width (GW) for Valmet FS5 of the fibre types. Asterisks (*) indicate that there is no statistical difference of a specific fibre trait with different analysers (α=0.05).

Fibre type	FibreShape Pro (µm)					QicPic (µm)				Valmet FS5 (µm)	
	GL	x_{10}	x_{50}	x_{90}	Aspect ratio	x_{10}	x_{50}	x_{90}	Aspect ratio	GL	GW
Virgin fibres, non-milled	1855.1*	384.5	1695.9	3457.9	0.27	36.1	260.0	870.1	0.22	1699*	34.3
Virgin fibres, milled	1687.8*	258.3	1497.4	3344.6	0.28	37.7	248.6	589.6	0.19	1513.5*	34.3
RF1-10, non-milled	1383.7	291.2	1156.1	2747.2	0.27	33.8	169.1	725.6	0.18	n.a.	n.a.
RF1-10, milled	1264.2*	268.0	1012.6	2592.6	0.27	36.4	306.7	901.3	0.20	1419*	35.2
RF1-16, non-milled	1296.8*	286.2	1095.7	2692.1	0.27	32.1	184.6	801.4	0.22	1322*	33.5
RF1-16, milled	1477.5*	318.7	1240.7	2930.8	0.27	34.3	195.8	702.4	0.23	1427*	34.6
RF2-10, non-milled	1218.0*	276.4	989.2	2434.1	0.27	49.5	408.9	877.0	0.19	1215*	35.6
RF2-10, milled	1129.5*	231.5	873.2	2376.0	0.30	44.5	255.8	722.9	0.17	1714*	34
RF2-16, non-milled	1210.0	280.3	1001.0	2398.3	0.29	41.9	256.0	737.7	0.15	n.a.	n.a.
RF2-16, milled	1287.7*	303.1	1091.4	2512.4	0.29	51.1	450.9	877.5	0.17	1412*	37.7

n. a. = "not analysed" due to the lack of sampling amount for RF1-10 (non-milled) and RF2-16 (non-milled) by Valmet FS5

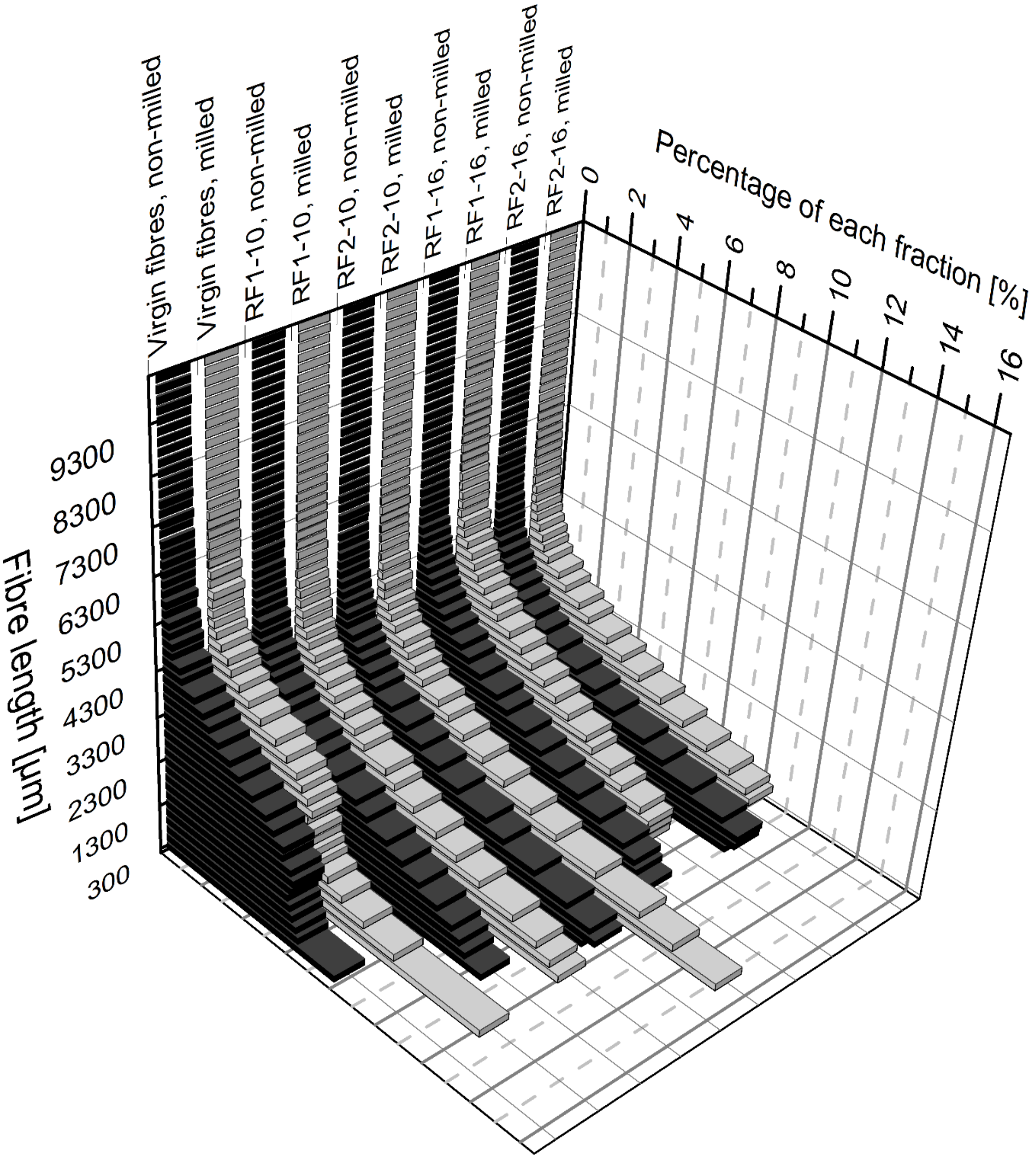

Figure 4.4: Percentage of each fraction of the length-weighted fibre length distribution for each fibre type based on FibreShape Pro. Non-milled samples are shown as dark curves, milled samples as light curves.

4.5.3 Comparison of dry and wet dispersion techniques

Wood fibres tend to absorb water; thus, swelling of these fibres is inevitable during fibre analysis in aqueous suspension. As radial and tangential swelling are 10 and 20 times larger than longitudinal swelling, methods based on aqueous suspensions generate lower aspect ratios compared to methods based on dry dispersion.

From a practical point of view, instruments with a wet dispersion method are particularly suitable for measuring pulp samples for paper making, as these are produced in an aqueous suspension. Therefore, drying would not lead to shrinkage of the fibres. On the other hand, thermo-mechanical pulp (TMP) fibres used for MDF production are more convenient for dry dispersion technique in order to not to get affected by the possible swelling during the analysis process. The investigations so far have shown that the three fibre analysers provide different absolute values for the fibre dimensions and that therefore the actual dimensions cannot be determined. However, the initial instruments give a very good representation of the relative dimensional changes that occur during the manufacturing and recycling process, regardless of whether it is based on a dry or wet dispersion process. Thus, both dispersion techniques can be applied for analysing the RF geometry.

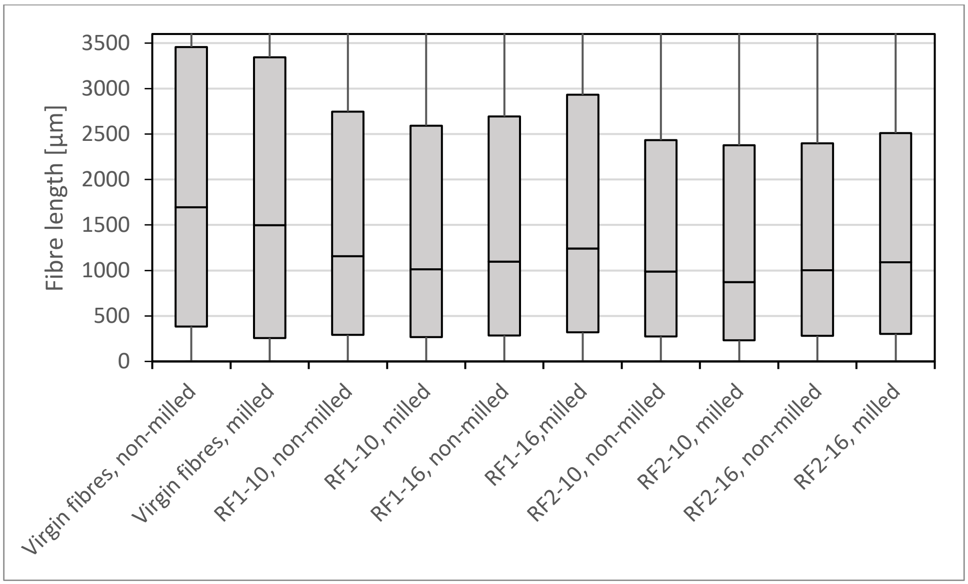

Figure 4.5: Box plot representation of geodesic fibre length (GL) based on FibreShape Pro.

4.6 Conclusions

The aim of this study was to analyse the impact of thermo-hydrolytic disintegration of waste MDF on the RF geometry. Three fibre analysers were used for the analysis and the results compared. The optical methods based on image analysis provided high resolutions in particle sizes. Yet, the results for length-based fibre length distribution (Q1) and GL show sizable differences between different analysers, suggesting that a relative comparison between different analysers is more feasible rather than an absolute comparison was. Nevertheless, results obtained from FibreShape Pro and Valmet FS5 were more consistent compared to results obtained from QICPIC. The obtained RF exhibited a fibre geometry similar to the initially applied fibres indicating that the geometry is principally maintained throughout the production and recycling process. Still, fibre shortening occurs due to MDF production and thermo-hydrolytic disintegration conditions.

Chapter 5

Drainage in a screw press and utilization of the recovered fibres after thermo-hydrolytic disintegration of waste fibreboards

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Authorship

Table 5.0: Contribution of the co-authors (as modified from Hunt 1991).

	Planning & Design	Executing & Data Collection	Data analysis & Interpretation	Literary input	Σ
F. Yağmur Bütün Buschalsky	25	25	25	25	**100**
Carsten Mai	15	0	15	15	**45**

F. Yağmur Bütün Buschalsky prepared the experimental design together with Carsten Mai. She performed the data collection, analysis and interpretation. Furthermore, she wrote the manuscript.

Carsten Mai prepared the experimental design together with F. Yağmur Bütün Buschalsky. He assisted in data interpretation and manuscript writing.

5.1 Abstract

A thermo-hydrolytic disintegration process qualifies as a promising option for recycling the waste MDF and preserving the fibrous morphology of the recovered lignocellulosic fibre material. This study aims to include a drainage process between the thermo-hydrolytic disintegration and the further utilization of the recovered fibres (RF) obtained using a screw press for removing the disintegration water (DW). In this context, the chemical properties of the RF (pH, nitrogen content, formaldehyde emissions) and the DW (pH, formaldehyde, reducing sugars and equivalents and nitrogen contents) were analysed. Moreover, the RF material was utilized to produce recycled MDF panels, solely containing the RF (100 %) and hence supplanting 50 % of the virgin fibres (VF). The recycled MDF portrayed significant reductions in the internal bond strength (IB), and flexural properties (MOR, MOE): in the case of MDF made from 100 % recycled fibres, about half the strength was reduced, and in the case of MDF made from 50 % recycled fibres, the strength was reduced by 20-25 %. The Thickness swelling (TS) of the recycled MDF panels was similar, while the water uptake (WA) was higher than that of the original MDF. The recycled MDF panels also exhibited a higher content of formaldehyde and emission. The findings recommend the application of a screw press process for prompter drainage of the RF and to utilize the RF obtained in combination with the VF to achieve adequate mechanical properties rather than using the RF separately for the manufacturing of the recycled MDF panels.

5.2 Keywords

fibreboards; MDF; wood-based panels; waste; thermo-hydrolytic disintegration; screw press; mechanical properties; physical properties; formaldehyde release

5.3 Introduction

Fibreboards are defined as a dry-formed panel product manufactured from ca. 80-90 % of fine lignocellulosic fibres are obtained from the thermo-mechanical pulping (TMP) from a refiner combined with a synthetic adhesive (generally urea-formaldehyde, UF resin). The global production of medium-density fibreboards (MDF) along with high-density fibreboards (HDF) reached a total of 98.6 million m^3 in 2018 (FAO 2019a). On completion of their service life, these panels turn into waste, and a majority are deposited in landfills or are burned. In Europe, waste wood materials consisting of more than 5 % of organic materials have been prohibited from landfilling since 2005 (Kharazipour and Kües 2007). Recycling of the MDF wastes presents an opportunity for extending the application of wood resources, reduction in the consumption of new resources, energy, landfilling as well as the expenses avoided through purchase/disposal fees, and helps in the creation of “green” jobs (Bratkovich et al. 2009). The thermo-hydrolytic disintegration process would be a promising option to preserve the fibrous morphology of the recovered lignocellulosic fibre material during the release of the fibres from the thermosetting resin matrix. Thermo-hydrolytic disintegration processes involve the application of liquid water, steam, heat and occasionally pressure for cleaving the existing adhesive bonds in waste MDF (Kharazipour and Kües 2007). Toward the end of the disintegration process, recovered fibres (RF) and a resin-water mixture, termed disintegration water (DW), are obtained. The implementation of an appropriate de-watering process for separating the RF from the DW is an important aspect for reducing the drying time of the RF.

The effects of using RF on the properties of the newly produced MDF after repeated thermo-hydrolytic disintegration processes have been investigated (Bütün Buschalsky and Mai 2021). This study aims to implement a drainage process between the previously researched thermo-hydrolytic disintegration process and the further utilization of the obtained RF using a screw press for removing the DW. In this context, the analysis of the chemical and morphological properties of the RF and the chemical properties of the DW was made. Moreover, the RF material is utilized for producing recycled MDF panels solely containing the RF (100 %) and supplanting 50 % of these by virgin fibres. The paper also explains the physico-mechanical as well as chemical (such as formaldehyde release) properties of these MDF panels.

5.4 Material and Methods

5.4.1 Materials

Three types of uncoated industrial fibreboards (medium-density fibreboard-MDF and high-density fibreboard-HDF) having different densities, thicknesses, as well as binder types and amounts, were used for the thermo-hydrolytic disintegration process (Table 5.1 and Figure 5.1). Moreover, the nitrogen content (NC) of the panels was determined using the Kjeldahl method as described previously (Bütün et al. 2019). The UF content of the MDF panels was calculated keeping in mind that the UF resin contained roughly 30 % nitrogen, neglecting the very low NC of the fibres (<0.1 %) used for producing the panels (Bütün Buschalsky and Mai 2021) (Table 5.1). However, due to the complexity in formulation, the UF content for Board-1 could not be calculated from the nitrogen content alone.

Table 5.1: Properties of the uncoated industrial fibreboards.

	Fibreboard properties	Board-1	Board-2	Board-3
	Fibreboard type	MDF	MDF	HDF
	Density (kg m^{-3})	792	739	862
	Thickness (mm)	6.0	19.0	6.6
Binder type	Urea formaldehyde (UF) amount (%)	5.5	9.9	14.7
	Melamine urea formaldehyde amount (%)	10.5	0	0
	Analysed nitrogen content (%)	6.0	3.6	5.0
	Calculated UF content (%)	n.c.	11.5	16.0

n.c. = "not calculated".

Figure 5.1: Uncoated industrial fibreboards used for the thermo-hydrolytic disintegration process.

5.4.2 Thermo-hydrolytic disintegration of waste fibreboards

Firstly, each fibreboard type was cut into pieces having dimensions of 25 × 25 mm^2 in cross-section by using a circular saw. The thermo-hydrolytic disintegration process was carried out separately for each type of fibreboard. Hence, for every disintegration, 9 kg of these fibreboard pieces were immersed in 30 liters of water at 95 °C in the autoclave Zirbus Z3 (Zirbus Technology GmbH, Bad Grund, Germany) for 30 min (Figure 5.2a). After every disintegration process, the recovered fibres, along with the disintegration water, were stored in barrels made of high-density polyethene and was sent to Maschinenfabrik Reinartz GmbH & Co. KG (Neuss, Germany).

5.4.3 Drainage of recovered fibres using a screw press

The excess water (disintegration water -DW) was removed using a screw press developed by Maschinenfabrik Reinartz GmbH & Co. KG (Neuss, Germany). Thus, the RF and DW were obtained separately (shown in Table 5.2 and Figure 5.2b). To determine the chemical and morphological properties, the RF was conditioned in a climate chamber at 20 °C with 65 % relative humidity.

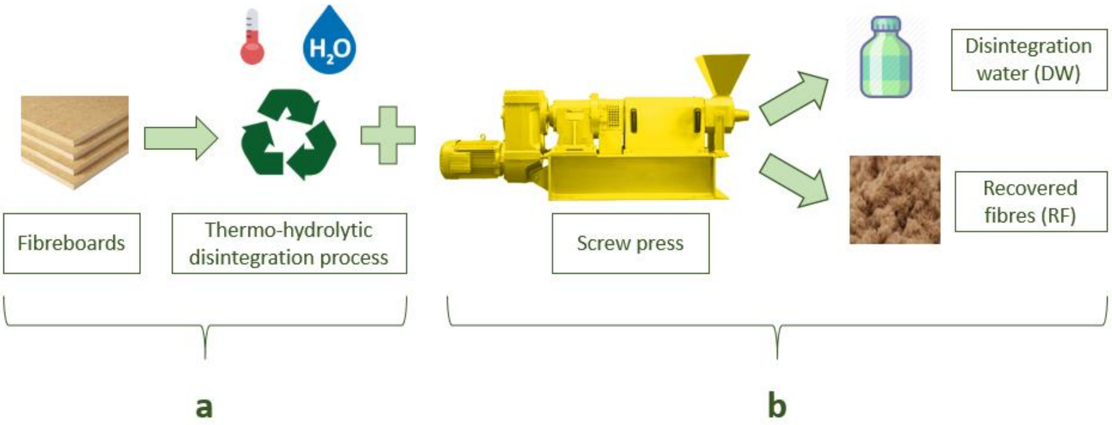

Figure 5.2: Schematic illustration of a) thermo-hydrolytic disintegration of waste fibreboards, b) drainage of recovered fibres using a screw press.

Table 5.2: Labelling system of the recovered fibres (RF) obtained and disintegration water (DW) samples after the disintegration processes.

Disintegrated fibreboard	Obtained recovered fibre	Obtained disintegration water
Board-1	RF1	DW1
Board-2	RF2	DW2
Board-3	RF3	DW3

5.4.4 Determination of the chemical properties of the recovered fibres (RF)

The pH value was measured after the extraction of fibres from cold water (4 specimens per fibre type); 2 g of sample material was placed in a 100 mL Erlenmeyer flask, and 60 mL of distilled water was added. Afterwards, the Erlenmeyer flask was placed on a shaker present at room temperature, and the measurements were made after periods of 50 min, 6 h, and 24 h, respectively, with the help of pH electrode InoLab Level 2 (WTW GmbH, Weilheim, Germany). Due to minor differences, only the average pH value was reported for each type of fibre. The nitrogen content was determined using the Kjeldahl method to assess the remaining UF resin content on the RF2 and RF3 after thermo-hydrolytic disintegration (4 specimens per fibre type), as previously described in (Bütün et al. 2019). Through the calculation of the remaining resin content on the RF, it was assumed that the UF resin contains roughly 30 % nitrogen. The proportion of initial contents of the binder, which had remained on the RF, was calculated by relating the NC of the RF to the NC of the respective MDF. Formaldehyde emissions from the fibre mass were determined based on the basic principle of the standard EN 717 3, which used approximately 2 g of the fibre material in a bleached tea bag (4 specimens per fibre type), and the values were recorded after 3 h of emission (DIN EN 717-3 1996). All the results were compared to industrially produced thermo-mechanical pulp (TMP) fibres (virgin fibres-VF) as a reference.

5.4.5 Chemical analysis of the DW

Analysis was made on the disintegration water samples for obtaining the pH value, including the formaldehyde content based on the acetylacetone method (DIN EN 120 1992) and nitrogen content based on the Kjeldahl method (4 specimens per DW sample) as described previously in (Bütün et al. 2019). The amount of reducing sugar equivalents in the DW was assessed based on the 3.5 dinitrosalicylic acids (DNSA) method (4 specimens per DW sample) (Miller 1959).

5.4.6 Production of the recycled MDF panels using RF2

Recovered fibres obtained after the disintegration of Board-2 were selected for manufacturing recycled MDF panels due to their compatibility with the thickness of the original board and resin content with the susceptible comparison of the previous research made by the authors. Recycled MDF panels (1 replica per panel) were manufactured using the RF2 obtained by the disintegration of the Board-2. Further disintegration of the RF2 was made in the hammer-mill VS1 N (Electra SAS, Poudenas, France) using a mesh size of 2.0 mm. Two different types of MDF panels containing 100 % and 50 % hammer-milled RF2 were produced and labelled as B100 and B50, respectively (Figure 5.3). The other 50 % of the formulation of the B50 was made up of industrially produced thermo-mechanical pulp (TMP) fibres (virgin fibres-VF). These were blended with the RF2 after hammer-milling as performed previously. The MDF panels were produced with 10% UF resin (Kaurit 350, BASF, Ludwigshafen, Germany) based on dry fibre mass and 2 % ammonium sulfate ($(NH_4)_2SO_4$) as a hardener based on the solid content of the resin. The resin and the hardener were pneumatically applied onto the fibres within a rotary drum using a spraying nozzle. The resinated fibre material was further hot-pressed (190 °C, 5 N mm^{-2}) for 15 s mm^{-1} to attain a target density of 700 kg m^{-3} using metal bar stops to reach a panel thickness of 16 mm.

Figure 5.3: Uncoated industrial Board-2 fibreboard and recycled MDF panels (B100 and B50) produced using recovered fibres obtained by the disintegration of Board-2 (RF2).

5.4.7 Determination of the physico-mechanical and chemical properties of the MDF panels

All the MDF panels and the respective cut specimens were conditioned at 20 °C having 65 % relative humidity until a constant mass was reached. The density of each individually cut MDF specimen was determined as per EN 323, and the overall density of the panel was calculated as the average of these samples (DIN EN 323 1993). In addition, the following properties were assessed: the moisture content (MC) according to EN 322 (36 specimens, 12 per panel), internal bond strength (IB) according to EN 319, the thickness swelling (TS) and water uptake (WU) after immersion in water for 24 h according to EN 317 while applying the sample dimensions of 50 × 50 × 16 mm^3 (4 specimens per Board-2 and three specimens per recycled panel), the flexural strength (MOR) and modulus of elasticity (MOE) according to EN 310 while applying the sample dimension of 370 × 50 × 16 mm^3 (two specimens per Board-2 and three specimens per recycled panel) (EN 310 1993; EN 317 1993; EN 319 1993; EN 322 1993). The Formaldehyde content was determined as per the EN 120, perforator method) (1 specimen per panel), while the formaldehyde emission of the boards was analysed according to EN 717-2 (gas analyser method (EN 717-2 1995)) (two specimens of Board-2 and one specimen per recycled panel) and flask method (EN 717-3 1996) with two specimens for every panel for 3 h and 24 h (EN 120 1992). The NC of the panels was determined using the Kjeldahl method (three specimens per Board-2 and two specimens for every recycled panel) as described previously in (Bütün et al. 2019). The binder content of the produced original and the recycled UF bonded MDF panels were calculated considering the UF resins contained roughly 30 % nitrogen when the very low NC of the virgin fibres were ignored (≤ 0.1 %) (Table 5.1 and Table

Table 5.3: Physico-mechanical and chemical properties of industrially produced B2, 100 % RF containing recycled B100, 50 % recovered fibres (RF) and 50 % virgin fibres containing recycled B50 MDF panels; N = specimen amount per panel, mean values (MV) ±standard deviation (SD).

MDF properties	Board-2			B100			B50		
MDF type and used fibre type	Industrially produced, 100% virgin fibres			Recycled, 100% RF2			Recycled, 50% RF2 and 50% VF		
	N	MV	SD	N	MV	SD	N	MV	SD
Number or replica panel	8	n/a	n/a	1	n/a	n/a	1	n/a	n/a
Density (kg m^{-3})	21	739.0	5.6	21	715.0	25.9	21	681.5	30.6
Thickness (mm)	n/a	19.0	n.c.	n/a	16.0	n.c.	n/a	16.0	n.c.
Moisture content (MC) (%)	12	7.8	≤0.1	12	7.2	≤0.1	12	6.8	≤0.1
Internal bond strength (IB) (N mm^{-2})	4	0.3	≤0.1	3	0.1	≤0.1	3	0.2	≤0.1
Flexural strength (N mm^{-2})	2	30.6	1.9	3	12.3	2.1	3	24.0	1.0
Modulus of elasticity (MOE) (N mm^{-2})	2	3735.3	50.4	3	1610.0	306.2	3	2566.0	254.8
Water uptake (%)	4	19.0	1.1	3	35.9	1.0	3	40.7	1.4
Thickness swelling (%)	4	10.0	1.6	3	10.1	0.1	3	8.9	0.2
Formaldehyde content - Perforator method (mg 100 g^{-1} o.d. board)	1	2.0	n/a	1	11.0	n/a	1	13.7	n/a
Formaldehyde emission - Gas analyser method (mg m^{-2} h^{-1})	2	2.5	≤0.1	1	10.1	n/a	1	10.5	n/a
Formaldehyde emission - Flask method (mg kg^{-1} o.d. board) 3 h	2	2.5	≤0.1	2	7.3	0.8	2	7.5	≤0.1
Formaldehyde emission - Flask method (mg kg^{-1} o.d. board) 24 h	n/a	n.a.	n.c.	2	104.7	5.7	2	116.1	6.7
Nitrogen content (%)	3	3.6	≤0.1	2	4.9	≤0.1	2	3.8	≤0.1
Calculated UF content (%)	n/a	11.5	n.c.	n/a	15.7	n.c.	n/a	12.2	n.c.

n/a = “not applicable”, n.c. = “not calculated”, n.a.=”not analysed”

5.3). All the aforementioned properties were also analysed for the Board-2 before being disintegrated using the thermo-hydrolytic process.

5.5 Results and Discussion

5.5.1 Chemical properties of the recovered fibres (RF)

The moisture content of the recovered fibres (RF) was as low as 20 %, indicating the effectiveness of the drainage process. As the moisture content was well below the fibre saturation point of coniferous wood such as spruce, it could be assumed that the degradation of the cell wall occurs or that the fibres were lost during processing. As bound water is held by intermolecular attraction within cell walls, this water cannot easily be removed unless the cell walls are broken (Glass and Zelinka 2010). The pH value of virgin fibres (VF) amounted to 4.0, while those of the recycling fibres (RF) ranged between 5.5 and 6.3 (Table 5.4). The higher pH of the RF may be attributed to the UF and the MUF resin itself, including the alkaline ammonia formed as a result of degradation from the resins during disintegration. Under acidic conditions in an aqueous environment, ammonia forms ammonium (Roffael and Hüster 2012; Wan et al. 2014), which results in the pH increment of the RF samples.

While the nitrogen content (NC) of the VF used for manufacturing the B50 was negligible (≤ 0.1 %), the NC of the recovered fibres was as high as 2.8 %, 1.7 % and 3.3 % for RF1, RF2 and RF3, respectively (Table 5.4). Thus, the resulting calculated binder content was 5.4 % and 10.6 % for RF2 and RF3, respectively (Table 5.4). Calculations were made on the amounts of initial binder contents found on the RF2 and RF3. The results revealed that the fibreboards possessed a relatively high tendency to be hydrolysed. Furthermore, 46.6 %, 47.0 % and 66.2 % of the initial binder content could still be found on the RF1, RF2 and RF3, respectively, implying that roughly half of the initial binder content was hydrolysed and dissolved in the DW samples while an insignificant amount evaporated during the processes of disintegration and drainage or even both. Due to the complexity in the formulation, the proportions of the UF and MUF for RF1 cannot be calculated using the nitrogen content alone (Table 5.4).

Formaldehyde emissions of the VF after 3 h were 5.6 and 9.1 mg per kg of the oven-dried (o.d.) fibres for the non-milled and hammer-milled samples, respectively (Table 5.4). As explained above, for NC, due to the remaining binder on the RF samples, formaldehyde emissions of the RF were 4.2 times higher than that of VF, on average. Thus, RF3, which

Table 5.4: Chemical properties of the virgin fibres (VF) and recovered fibres (RF), mean values (MV) ± standard deviation (SD).

Chemical properties	Virgin fibres (VF)				RF1		RF2		RF3	
	Non-milled		Hammer-milled							
	MV	SD	MV	SD	MV	SD	MV	SD	MV	SD
Moisture content after drainage (%)	n/a	n/a	n/a	n/a	20.1	n.c.	22.0	n.c.	20.7	n.c.
Moisture content after conditioning (%)	7.1	≤0.1	8.9	≤0.1	17.9	2.1	11.0	0.3	11.4	0.8
pH (50 min, 6 and 24 h)	4.2	≤0.1	4.2	≤0.1	6.3	0.1	5.5	0.1	6.2	0.1
Nitrogen content (%)	0.1	≤0.1	0.1	≤0.1	2.8	≤0.1	1.7	≤0.1	3.3	≤0.1
Calculated UF content (%)	n.c.	n.c.	n.c.	n.c.	n.c.	n.c.	5.4	n.c.	10.6	n.c.
Amount of initial binder content remained on the fibres (%)	n/a	n.c.	n/a	n.c.	46.6	n.c.	47.0	n.c.	66.2	n.c.
Formaldehyde emission - Flask method (mg/kg o.d. fibre) (3 h)	5.6	0.2	9.1	0.07	57.2	0.9	29.6	5.8	36.6	7.5

n/a = “not analysed”, n.c. = “not calculated”.

contains higher amounts of NC (3.3 %), releases higher formaldehyde (36.6 mg per kg o.d. fibre) compared to the RF2 (29.6 mg per kg o.d. fibre). The formaldehyde release was not found to be proportional to the NC content. Thus, RF1 exhibited a slightly lower NC (2.8 %) compared to RF3 but released more amounts of formaldehyde (57.2 mg per kg o.d. fibre), although the binder contained MUF resin which is considered to be more stable during hydrolysis.

5.5.2 Chemical properties of the disintegration water (DW)

The thermo-hydrolytic disintegration water samples obtained from the MDF panels exhibited higher pH values when compared to demineralized water (Table 5.5). This is attributed to the formation of ammonia (NH_3), which reacts with water to form ammonium hydroxide (NH_4OH) (Moezzipour et al. 2017). The amount of formaldehyde present in the DW after the disintegration processes was 1618.8 mg L^{-1}, 813.6 mg L^{-1}, and 1385.3 mg L^{-1} for DW1, DW2 and DW3, respectively. Significant levels of reducing sugar equivalents are detected in the DW samples (shown in Table 5.5), which may be products having a cleavage, mainly consisting of hemicelluloses released during the production of VF and/or the thermo-hydrolysis of the MDF panels. The nitrogen content of the DW1, DW2 and DW3 was found to be 0.8 %, 1.0 % and 1.2 %, respectively (Table 5.5). On comparing the chemical properties of the DW samples to that of a previous study made by the authors (F.Y.B.B., unpublished data 2020), the screw press process was seen to have an alteration; the pH values were more alkaline, while the DW contained more dissolved UF (higher NC of the samples). Thus, higher formaldehyde content and fewer equivalents for reducing sugars were seen. A possible explanation for this alteration could be due to typically volatile ammonia, a by-product of the UF-resin breakdown, that would have been captured and not released, as the obtained recovered fibres together with the disintegration water were stored in barrels made of high-density polyethylene and were sent to Maschinenfabrik Reinartz GmbH & Co. KG (Neuss, Germany).

Table 5.5: Chemical properties of the disintegration water (DW) obtained after the thermo-hydrolytic disintegration processes compared to the demineralised water mean values (MV) ± standard deviation (SD).

Chemical properties	DW1		DW2		DW3		Demineralised water
	MV	SD	MV	SD	MV	SD	
pH	5.7	≤0.1	5.4	≤0.1	6.4	≤0.1	5.2
Formaldehyde content (mg l^{-1})	1618.8	0.4	813.6	≤0.1	1385.3	≤0.1	n/a.
Reducing sugars and equivalents (mg l^{-1})	5.7	1.3	1.2	0.2	7.8	0.3	0.2
Nitrogen content (%)	0.8	≤0.1	1.0	≤0.1	1.2	≤0.1	≤0.1
Calculated UF content (%)	n.c.	n.c.	3.2	n.c.	3.8	n.c.	n.c.

n/ a. = "not analysed", n. c. ="not calculated".

5.5.3 Physico-mechanical and chemical properties of the MDF panels

It was seen that when 100 % of the recovered fibres containing recycled MDF (B100) exhibited a slightly higher density, the B50 exhibited a lower density when compared to the target density (see Table 5.3). The recycled MDF containing 100 % RF (B100) had an IB, which was nearly half as high as that of the original MDF (Board-2). The IB of the other recycled MDF (B50) was 28.5 % lower than the original MDF, even though its final density was lower by 7.8 %. These results are contrary to the previous studies made by the authors (Bütün Buschalsky and Mai 2021) , in which increased IB results were seen when 100 % of the RF was used for the production of the recycled MDF. In addition, in the present study, the amount of binder content remaining on the RF was roughly 20 % higher than the study mentioned above, and the fibres were severely shortened. Thus, the reduced IB of the B100 panel could be due to the over proportional consumption of the adhesive by fines and loss of strength of the RF (Roffael et al. 2016). The flexural strength (MOR) of the recycled MDF, B100 and B50 was lower by 59.8 % and 21.5 % than the original MDF panel, respectively. Similarly, the modulus of elasticity (MOE) of the recycled MDF panels was also reduced by the same range as the MOR compared to the original MDF. As for the IB, it can be attributed to the shortened fibre length due to the thermo-hydrolytic disintegration conditions, by subsequent hammer-milling and through the severe drainage process in the screw press. The properties related to the strength of these recycled MDF were significantly lower compared to those of the previous studies, which used the de-watering of the RF within a spin dryer (Bütün Buschalsky and Mai 2021). In the MDF, the length of the fibre particularly determines the flexural strength and stiffness of the boards (Steffen et al. 2001; Benthien et al. 2014a). This indicates that a severe shortening of the RF occurs in the screw press. Another possible explanation for these reductions might be that the flexibility of the RF was reduced due to the thermo-hydrolytic disintegration process, making the RF more brittle. Brittleness, in turn, could lead to the shortening of fibre under the mechanical impact.

Compared to the original MDF panel (Board-2), the recycled MDF containing 50 % of RF displayed an 11 % decrease in the thickness swelling (TS), while the other recycled MDF (B100) showed no difference. The latter result is surprising since the IB of the B100 variant was reduced by about 50 %, and the TS generally decreases with the increase in the bond quality. Water uptake (WU) after 24 h immersion, however, was increased with the average ca, two times for the recycled panels. When compared to two recycled panels, the slightly lower WU of B100 can be attributed to the reduced wettability of the RF due to the presence of the remaining resin. In addition, hydrophilic sugars derived from the degradation of hemicelluloses might be washed out during recycling. These sugars might absorb water, particularly in the virgin boards. Thus, the NC and the calculated UF content of the B100 was higher. B100 is supposed to contain ca. nitrogen content of 3 % derived from the utilized 10 % UF binder and 1.7 % nitrogen is derived from the used RF. The obtained result of 4.9 % NC is under this assumption, and the same applies accordingly to B50.

The formaldehyde content (2.0 mg per 100 g o.d. board) of the original MDF panel was far below the upper limit value of 8 mg per 100 g o.d. board, as per the standard EN 120 (EN 120 1992). The recycled MDF, however, exhibited a very high amount of formaldehyde, which might have been derived from both the new UF resins that were used for producing the recycled MDF and the remaining binder on the RF.

Similarly, formaldehyde emissions of the recycled MDF panels were significantly higher than of the original MDF panels irrespective of the employed method (gas analyser and flask). No difference was noted between the two recycled MDF panel types. In a previous study made by the authors (Bütün Buschalsky and Mai 2021), lower formaldehyde emissions were noted from the recycled boards compared to the virgin panels, which was attributed to scavenging of the formaldehyde due to reaction with urea, ammonia, and oligomeric decay products of the UF resin. This would result in lower formaldehyde emissions for B100 compared to B50.

However, it should also be noted that the limitations of the laboratory-scale production of the recycled MDF panels could cause some inconsistent physico-mechanical and chemical results when compared to the industrially produced Board-2.

5.6 Conclusions

The present study aims to implement a drainage process based on the application of a screw press, which occurs in between the thermo-hydrolytic disintegration process and the further utilization of the obtained RF (e.g., for production of new panels). Compared to the previous study, which applied de-watering in a spin dryer, the screw press process produces dryer fibres but affects the chemical properties of RF as well as the DW differently. One of the most significant findings of this study is that the recycled MDF panels showed a decrease in mechanical properties compared to the original MDF panels. Although improvement in these properties was seen with the mixing of RF with VF, the strength of the panels containing only the VF were not received. Altogether, drainage in a screw press is very efficient; however, pressing too hard to achieve a low moisture content would result in severe fibre damage. Thus, either the pressing force must be reduced, or the RF should be used in combination with the VF to obtain better mechanical properties for the recycled MDF panels.

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Competing Interests

The authors have declared that no competing interests exit.

Chapter 6

Recovering fibres from fibreboards for wood polymer composites (WPC) production

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Authorship

Table 6.0: Contribution of co-authors (as modified from Hunt 1991).

	Planning & Design	Executing & Data Collection	Data analysis & Interpretation	Literary input	Σ
F. Yağmur Bütün Buschalsky	15	20	15	20	**70**
Aaron K. Mayer	10	20	10	5	**45**
Kolja Ostendorf	10	20	10	5	**45**
Ole-Elias Z. Gröne	10	20	10	5	**45**
Kim C. Krause	15	5	5	5	**30**
Christian Schöpper	10	0	5	0	**15**
Oliver Mertens	5	0	5	0	**10**
Andreas Krause	5	0	5	5	**15**
Carsten Mai	10	0	15	15	**40**

F. Yağmur Bütün Buschalsky prepared the experimental design together with Aaron K. Mayer, Kolja Ostendorf, Ole-Elias Z. Gröne, Kim C. Krause, Christian Schöpper, Oliver Mertens, Andreas Krause and Carsten Mai. Furthermore, she performed data collection together with Aaron K. Mayer, Kolja Ostendorf, Ole-Elias Z. Gröne and Kim C. Krause. She analysed and interpreted the data and wrote the manuscript.

Aaron K. Mayer prepared the experimental design together with F. Yağmur Bütün Buschalsky, Kolja Ostendorf, Ole-Elias Z. Gröne, Kim C. Krause, Christian Schöpper, Oliver Mertens, Andreas Krause and Carsten Mai. Furthermore, he performed data collection together with F. Yağmur Bütün Buschalsky, Kolja Ostendorf, Ole-Elias Z. Gröne and Kim C. Krause and supported data interpretation and manuscript writing.

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6.1 Abstract

In most countries, fibreboards are not recovered after utilisation but burned for energy production. This study aims at recovering fibres from industrial fibreboards and reusing them as reinforcement elements in wood polymer composites (WPC). Recovered fibre (RF) material was generated by the thermo-hydrolytic disintegration of medium and high density fibreboards bonded with urea-formaldehyde resin. Various formulations of RF and polypropylene were used with or without the addition of the coupling agent to manufacture WPC using a co-rotating extruder. Test specimens were produced via injection moulding whereby those containing 'virgin' fibres served as a reference with respect to mechanical and physical properties. WPC formulations containing RF and 'virgin' fibres exhibited similar results, but composites containing RF exhibited improved mechanical and water-related properties, especially without coupling agent. The study indicates that recovered fibres are suitable to produce WPC with very similar physico-mechanical properties as those from 'virgin' fibres.

6.2 Keywords

cascade use; fibreboard; MDF; wood polymer composite; recycling; mechanical properties; physical properties; thermo-hydrolytic-disintegration

6.3 Introduction

Cascading use of biomass is a principle where products with higher value added are produced first and energy at a later stage of the production chain (Indufor 2013). It is the efficient utilization of resources by using residues and recycled materials to extend the total biomass availability within a given system (Sokka et al. 2015). A cascade use of wood is defined as multiple use of the wood resources from trees by using residues, recycling (utilization in production) resources or recovered (collected after consumption) resources (Mantau 2012). The increased cascading use enhances resource efficiency.

Fibreboards are defined as a dry-formed panel product manufactured from lignocellulosic fibres combined with a synthetic resin or other suitable binders. Global fibreboard production has reached 105 million m^3 in the year 2015 (FAO 2016). This growth induces waste fibreboard generation which is about 40.5 million m^3 annually (Irle et al. 2018). After service life, waste fibreboards are burned for energy production in most countries (BAV 2015). One of the possibilities of re-using these waste fibreboards is processing them via a thermo-hydrolytic disintegration process and then producing Wood Polymer Composites (WPC).

Michanickl and Boehmke (1998) patented a thermo-hydrolytic treatment to disintegrate wood-based panels. The treatment is used for recovering particles and fibres from residues of items such as old pieces of furniture, production residues, water and other wood-containing materials bonded with hydrolysable resins. Furthermore, Roffael and Dix (1996) developed a technique based on chemical pulping of waste particle and fibreboards by the aqueous alkaline solution. MDF Recovery Ltd together with the BioComposites Centre work on developing a technology to recover wood fibre from waste medium density fibreboards (MDF) for generating a new raw material source for the industry with close to market approach (BioComposite Centre 2015).

Wood-Polymer Composites (WPC) are composites that exhibit thermoplastic processing behaviour and consist of varying contents of wood, plastics and additives, which are processed by thermoplastic shape-forming techniques such as extrusion, injection moulding, roto-moulding or compression moulding (Vogt et al. 2006; Teuber 2016). The market demand for WPC has rapidly increased as an important family of engineering materials (Zimmermann and Zattera 2013). The increasing acceptance of WPC is due to their low moisture absorption, dimensional stability, and resistance to biological attack, combination of high specific stiffness and strength, relatively low abrasion during processing, low density and mouldability (Valente et al. 2011), as well as the design and the appearance of the final products. Natural organic fillers present lower density, abrasiveness and cost, they are renewable and biodegradable (Clemons 2002).

The primary objective of wood as a filler material is to increase the stiffness of WPC (Gozdecki et al. 2015). In general, wood as a filler material can be used in two different ways; wood fibres and wood flour. Wood flour is used as filler for plastic, which tends to increase the stiffness of the composite but does not improve its strength. From a technical point of view, wood fibres may contain single anatomical fibres and bundles, whole fibres or fibre fragments. Wood fibres can be obtained through mechanical (TMP – thermomechanical pulp) or chemical (bleached or unbleached pulps) processing (Mertens et al. 2017a). Wood fibres can be used to reinforce the composites with respect to both strength and stiffness (Stark and Rowlands 2003). Wood fibres differ from wood flour in that fibres are significantly longer than their widths, rather than particle like (Matuana and Stark 2014). The aspect ratio of fillers (length to width ratio) has a greater influence on mechanical properties of WPC than filler size (Stark and Rowlands 2003; Nourbakhsh et al. 2010; Gozdecki et al. 2011; Teuber 2016). Therefore, the aspect ratio plays an important role regarding the reinforcement of composites, and subsequently for determining the mechanical properties of the final product. Reinforcement occurs when the fibres have a minimum (critical) length l_c that allows stress transfer from the matrix to the fibre (Monette et al. 1993). Higher aspect ratio reportedly increases flexural and tensile strength and modulus, as well as unnotched impact bending (Stark and Berger 1997). That is why, the utilization of wood fibres instead of wood flour as reinforcement in WPC has gained interest in recent years (Lerche et al. 2014; Mertens et al. 2017b). Severe fibre length reduction, however, occurs during the composite preparation because of the chosen raw material feeding method, low polymer melt flow rate and increasing screw speed in the twin screw extruder as well as high shear forces during the injection moulding process (Teuber et al. 2016a, b).

As the wood component of WPC is hydrophilic and the plastic is hydrophobic, a compatibilizer is often used to improve interfacial bonding of the two different phases (Caulfield et al. 2005). The function of the compatibilizer is to promote covalent bonds and/or secondary acid-base links or hydrogen bonding between wood fiber and PP (Zimmermann and Zattera 2013). Maleic anhydride-grafted polypropylene (MAPP) is one of the most widely used compatibilizer in WPC manufacturing due to its low cost and fair efficiency (Li and He 2004; Adhikary et al. 2008; Dányádi et al. 2010). MAPP is thought to work by two different mechanisms. First, the anhydride reacts with a cell wall polymer hydroxyl group to form an ester bond, then the PP attaches to the anhydride intertangles into the PP network in the melt(Caulfield et al. 2005). Numerous studies reported improvements in tensile strength, elongation in break and unnotched impact bending (Oksman and Clemons 1998).

Recovered fibres (RF) obtained after the thermo-hydrolytic disintegration of fibreboards might be used as filler and/or reinforcing agent for thermoplastic polymers to produce WPC. Such utilization can be a good intermediate in the cascade chain (Migneault et al. 2014) and provide more efficient usage of the wood resources.

The objective of this study is to evaluate the mechanical and physical properties of WPC containing RF and compare it to identically produced composites containing ‘virgin’ MDF fibres directly produced from wood. The respective composites were produced with and without MAPP as coupling agent.

6.4 Material and Methods

6.4.1 Disintegration of the fibreboards

Uncoated industrial fibreboards were obtained from Glunz AG (Meppen, Germany). The MDF exhibited a thickness of 19 mm, raw density of 0.736 g cm^{-3} and urea formaldehyde (UF) resin content of 9.9 %. The respective values for the high-density fibreboards (HDF) were 7.4 mm thickness, raw density of 0.856 g cm^{-3} and UF resin content of 15.1 %. For each disintegration process, 2.00 kg of MDF/HDF pieces (50 x 50 mm in cross-section) were immersed in 35 L water and heated to 95 °C for 8 min (MDF) or 11 min (HDF) in an autoclave. After drying of the RF to 5 % moisture content at 70 °C, they were further disintegrated in the hammer-mill VS1 N (Electra SAS, Poudenas, France), using an aperture/mesh size of 2.00 mm.

6.4.2 Composite preparation

The isotactic polypropylene (PP) Sabic 575P (Sabic, Riyad, Saudi Arabia) was used as polymer matrix; its density was 0.905 g cm^{-3}, and its melt flow index was 11 g 10 min^{-1} (230 °C/2.16 kg). MAPP Licocene 7452 TP (Clariant, Muttenz, Switzerland) was used as a coupling agent, its density was 0.930 g cm^{-3} and grafting level was 7 %. In addition to the RF, 90 % Scots pine (*Pinus sylvestris*) and 10 % radiata pine (*Pinus radiata*) fibres referred to as ‘virgin’ MDF fibres, were used as reference material after treated in the hammer-mill VS1 N (Electra SAS, Poudenas, France), using an aperture/mesh size of 2.00 mm. Oven-dried (moisture content ≤ 5 %) recovered and ‘virgin’ fibres, PP and MAPP were compounded for each formulation (Table 6.1). A co-rotating twin-screw extruder MICRO27GL/GG40D (Leistritz, Nürnberg, Germany) was used for compounding. The average temperature from zone 1 to zone 9 were 180 °C, 175 °C, 175 °C, 175 °C, 170 °C, 165 °C, 160 °C, 153 °C and 145 °C, respectively. The average melting temperature was kept around 140 °C, with an average screw speed of 130 rev min^{-1} and pelletizer speed of 240 rev min^{-1}. Because of the low fibre bulk density, bridge building of the fibres occurred at the feeder inlet; therefore the fibres were manually forced towards the feeder.

Table 6.1: WPC formulations (percent by weight).

Homogenous groups (HG)	Wood fibre content (%)	Polypropylene (PP) content (%)	Maleic anhydride grafted polypropylene (MAPP) content (%)
A	60	40	0
B	40	60	0
C	20	80	0
D	60	37	3
E	40	57	3
F	20	77	3

6.4.3 Injection moulding of the compounds

The WPC compounds were further processed in a grinding mill SM 2000 (Retsch, Düsseldorf, Germany) with 4.00 mm aperture/mesh size. Thereafter, all compounded formulations were injection moulded using ALLROUNDER 420 C (Arburg, Loßburg, Germany) at a temperature of 170 °C with pressure ranges between 1500 and 2500 bar and average injection pressure time of 1.2 s.

6.4.4 Dynamic image analysis of the fibres

Fibre size and shape were determined after hammer-milling and prior to compounding using dynamic image analysis system QICPIC combined with dry dispersion unit RODOS (Sympatec GmbH, Clausthal-Zellerfeld, Germany). In the device, the wood fibre particles were dispersed in an accelerated air jet and a high speed camera took pictures of the particle stream with a resolution of 10 and 20 µm. Measuring particle size and shape from the projected particle area of binary pictures and calculating size and shape distributions were done by the software WINDOX (Sympatec GmbH, Clausthal-Zellerfeld, Germany) (Teuber et al. 2016b). Fibre length was characterized with respect to the median (x_{50}) and quartiles (x_{25} and x_{75}) of the length-based particle length distribution (Q1). The number of analysed particle per measurement varied between 215,000 and 249,000, and 3 measurements were consecutively repeated for each fibre type. The WINDOX software provides the length as one parameter for describing the particle shape. Thereby, it is defined as the aspect ratio of the particle (length to width ratio).

6.4.5 Characterisation of mechanical and physical properties

Rod-shaped specimens (80 x 10 x 4 mm) were used to determine bending properties, unnotched impact bending (Charpy) and water absorption according to the standards DIN EN ISO 178 (2006), DIN EN ISO 179 (2006) and DIN EN ISO 15534-1(2007), respectively. In parallel with the water absorption test, volume change of the WPC was determined using the same specimens. The water absorption and the volume change of the specimens were assessed by weighing and volume determination after 0, 1, 2, 7, 14, 21 and 28 days of immersion in water at 21 ± 2 °C and calculated according to Eq. 1 and Eq. 2, respectively. Tensile properties were determined with dumbbell-shaped specimens (170 x 10 x 4 mm) according to DIN ISO EN 527 (2003). Ten replicates were used for each test.

$$\text{Moisture absorption}(\%)=\frac{W_t\text{-}W_o}{W_o}*100 \qquad (1)$$

Where W_t is the specimen weight after a given immersion time in water and W_o is the weight of the specimen after conditioning at 21 ± 2 °C and 65 % relative humidity.

$$\text{Volume change }(\%)=\frac{V_t\text{-}V_0}{V_0}*100 \qquad (2)$$

Where V_t is the specimen volume after a given immersion time in water and V_o is the volume of the specimen after conditioning at 21 ± 2 °C and 65 % relative humidity.

6.4.6 Statistical analysis

The obtained data were statistically analysed using the RStudio version 3.4.3 (RStudio: Integrated Development for R Inc., Boston, Massachusetts, USA). The two-way analysis of variance (ANOVA) was conducted at a level of significance of α=0.05 to determine the significance of the effect of RF on WPC mechanical and physical properties. Tukey's HSD (honest significant difference) test was applied to evaluate the statistical significance between mean values of the WPC properties with different fibre types produced with and without MAPP.

6.5 Results and Discussion

6.5.1 Mechanical properties

In general, WPC containing recovered and 'virgin' fibres exhibited very comparable strength properties, but the values for WPC with RF were always higher (except for the unnotched impact bending at 20 % wood fibre content). This is especially true for high fibre content. (Tables 6.2 and 6.3). The addition of MAPP enhanced the mechanical properties for all formulations irrespective of the fibre type. This is attributed to a better, probably covalent bonding between fibres and PP matrix (Lu et al. 2000; Stark and Rowlands 2003; Gozdecki et al. 2015).

Table 6.2: Tensile strength and tensile modulus of the WPC with and without MAPP depending on the fibre type and the fibre content (HG); n=10, mean values (MV) ± standard deviation (SD).

		Tensile strength (MPa)						Tensile modulus (GPa)					
	MAPP	Virgin		RF1		RF2		Virgin		RF1		RF2	
HG	(%)	MV	SD	MV	SD	MV	SD	MV	SD	MV	SD	MV	SD
A	0	20.5	±0.1	23.9	±0.1	22.6	±0.1	5.0	±0.0	5.1	±0.0	5.1	±0.0
B	0	25.6	±0.2	27.9	±0.1	26.9	±0.1	4.1	±0.0	3.9	±0.0	3.9	±0.0
C	0	27.9	±0.1	28.2	±0.1	28.3	±0.1	2.8	±0.0	2.6	±0.0	2.7	±0.0
D	3	49.0	±0.6	49.7	±0.9	49.5	±0.7	5.9	±0.0	5.9	±0.0	5.9	±0.0
E	3	43.3	±0.1	44.2	±0.4	45.6	±0.2	4.2	±0.0	4.2	±0.0	4.4	±0.0
F	3	35.8	±0.2	36.1	±0.1	37.4	±0.3	2.7	±0.0	2.8	±0.0	2.9	±0.0

The results indicate that the disintegration process does not negatively affect the mechanical properties of the WPC compared to those containing ‘virgin’ fibres. Statistically, the RF1 fibres were somewhat smaller than the ‘virgin’ fibres, but the RF2 fibre length was in the same range as the latter (Table 6.4). The fibre content and the addition of MAPP have different effects on the mechanical properties of the WPC specimens.

Wood fibres have reinforcing effects on composites. Mechanical properties of WPC depend on numerous parameters including the fibre content and fibre length (Mertens et al. 2017a). Increasing fibre length lead to improved mechanical properties of WPC with and without MAPP (Stark and Rowlands 2003; Migneault et al. 2009). On the other hand, strength properties such as tensile and flexural strength decrease with increasing wood fibre content in the absence of MAPP (Yam et al. 1990) but increased when MAPP was added (Gozdecki et al. 2015).

It has previously been shown, however, that WPC production processes such as extrusion cause severe shortening of the fibres (Wolcott and Englund 1999; Migneault et al. 2009). Fibre shortening and resulting decrease in aspect ratio during disintegration of fibreboards is therefore minor to that which occurs during WPC production.

Generally mechanical properties increase with the compatibility of the fibre surface and the matrix (Caulfield et al. 2005; Zimmermann and Zattera 2013). The mechanical properties of the filler-reinforced composites are mainly influenced by the interfacial adhesion between

Table 6.3: Flexural strength, flexural modulus and unnotched impact bending of the WPC with and without MAPP depending on the fibre type and the fibre content (HG); n=10, mean values (MV) ± standard deviation (SD).

		Flexural strength (MPa)						Flexural modulus (GPa)						Unnotched impact bending (kJ m^{-2})					
	MAPP	Native		RF1		RF2		Native		RF1		RF2		Native		RF1		RF2	
HG	(%)	MV	SD	MV	SD	MV	SD	MV	SD	MV	SD	MV	SD	MV	SD	MV	SD	MV	SD
A	0	37.2	±0.0	43.8	±0.8	41.0	±0.3	5.6	±0.0	5.6	±0.1	5.7	±0.0	6.6	±0.9	7.1	±1.4	7.6	±1.0
B	0	42.8	±0.5	48.4	±0.8	45.5	±1.0	3.8	±0.0	4.0	±0.1	3.7	±0.1	10.5	±1.0	11.7	±1.8	13.2	±1.5
C	0	41.6	±0.6	47.8	±0.5	44.1	±1.1	2.2	±0.0	2.6	±0.3	2.3	±0.0	20.3	±1.2	18.3	±4.3	18.0	±3.6
D	3	81.2	±1.8	84.6	±1.1	85.0	±0.9	6.0	±0.0	6.4	±0.1	6.1	±0.1	13.6	±1.3	15.3	±1.6	15.1	±1.2
E	3	68.3	±0.8	72.5	±1.2	72.9	±1.0	3.6	±0.2	4.2	±0.0	4.0	±0.1	18.5	±1.4	19.2	±2.6	17.6	±2.6
F	3	49.6	±0.8	57.6	±0.4	53.2	±1.4	2.1	±0.0	2.7	±0.0	2.4	±0.1	25.3	±1.6	22.5	±3.7	24.8	±3.6

the fillers and the polymer matrix (Gwon et al. 2010). Thus, MAPP enables a transfer of loads from the PP matrix to the fibres (Zaini et al. 1996). Better adhesion between the polymer and the reinforcement element provides a higher mechanical strength of the composites due to the transferred high wood fibre strength to the PP matrix.

Accordingly our results show that, without a coupling agent, increasing wood fibre content in the mixing ratio slightly decreased the tensile strength irrespective of the fibre type (Table 6.2). At 60 % and 40 % fibre content, WPC containing RF showed significantly higher tensile strength values than those containing ‘virgin’ fibres. Adding MAPP, however, steadily increased the tensile strength with increasing fibre content for all the fibre types. WPC containing recovered and ‘virgin’ fibres with MAPP did not exhibit clear differences in tensile strength.

The differences in flexural strength between the fibre types were greater than those in tensile strength. These differences were greater without coupling agent (Table 6.3). WPC containing RF revealed 14 % higher flexural strength than those containing ‘virgin’ fibres. Differences regarding the fibre types decreased with decreasing fibre content of the composites. After adding MAPP, flexural strength was about 36 % higher at 60 % than at 20 % fibre content (Table 6.3), while tensile strength was some 26 % higher (Table 6.2) for all fibre types. This might be explained by the higher compression strength of composites with higher fibre content, due to the increasing density of the WPC; the true density of wood amounts to approx. 1.500 g cm^{-3} (Stamm 1929) while that of PP to approx. 0.905 g cm^{-3}.

For all fibre types, with and without a coupling agent, tensile and flexural moduli increased continuously with increasing fibre content from 20 % to 60 % (Tables 6.2 and 6.3). WPC from recovered and ‘virgin’ fibres did not show clear differences. Addition of MAPP just slightly increased the flexural and tensile moduli. Higher moduli values with increasing wood fibre content are due to the elastic modulus of wood. It is higher than that of the polymer especially in the grain direction (Gozdecki et al. 2015). Thus, the presence of fibres in the compound is known to enhance the modulus of elasticity, while the matrix polymer determines the flexibility and the ability of the composite to deform (Stark and Rowlands 2003; Gozdecki et al. 2015). Higher modulus values can be attributed to better compaction and fibre/matrix adhesion gained by injection moulding process (Mertens et al. 2017b).

WPC from RF exhibited clearly higher unnotched impact bending than those of ‘virgin’ fibres, particularly without MAPP (Table 6.3). At 60 % fibre content, unnotched impact bending of the composites containing RF1 and RF2 were 24 % and 29 % higher than those containing ‘virgin’ fibres. WPC with MAPP revealed lower differences with respect to the fibre type. Generally, unnotched impact bending decreased with increasing wood content in the mixing ratio both with and without MAPP irrespective of the fibre type. Without MAPP, unnotched impact bending decreased by approximately 64 %, while with MAPP it decreased by 38 %, when the fibre content increased from 20 to 60 %. Addition of MAPP clearly increased the unnotched impact bending by 21 %, 36 % and 54 % at 20 %, 40 % and 60 % fibre content, respectively. In respect to flexural and tensile strengths, the respective improvements in unnotched impact bending induced by MAPP were progressively higher with increasing wood content presumably because of better interfacial bonding between fibres and PP.

Unnotched impact bending indicates the amount of energy absorbed by the composite during fracture (Cantwell and Morton 1991). It is the sum of the energies required to break the sample. Wood fibres in the WPC formulations act as stress concentrators, thus decreasing the impact energy (Bledzki et al. 1998; Nabi Saheb and Jog 1999).

Higher fibre content strongly decreases the unnotched impact bending, which is given by the area below the stress-strain curve. This indicates that with high fibre content, the deformation of the specimens during a failure is much lower than that of specimens with low fibre content. Thus, the addition of fibres reduces the flexibility (pliability) of the WPC.

The results indicate that WPC containing RF exhibit higher tensile and flexural strength as well as impact bending, while the moduli of RF composites were the same as those from ‘virgin’ fibres. This indicates that all composites have the same stiffness. The slightly higher strength properties might be due to the better compatibility of the RFs’ surface with the PP matrix, probably caused by remaining UF adhesive on the fibre surface. PP (matrix material) or MAPP might better interact with the fibre surface, because the RFs’ surfaces are activated through disintegration (e.g. polarity is more similar to PP and PP better interacts with remaining adhesive) and/or because MAPP can interact (physically or even covalently) with remaining adhesive on the RF surface.

Another reason for the slightly higher strength of the RF composites might be an enhanced flexibility of the RF caused due to the influence of water at high temperature (95 °C) during the disintegration process.

6.5.2 Water absorption and volume change

The WPC specimens showed continuous water absorption over 28 days, regardless of the fibre type (Table 6.5). As expected, WPC formulations with higher wood content tended to absorb more water than those with a higher proportion of polymer, due to the hygroscopic chemical structure of the wood cell wall. Without MAPP, WPC containing RF absorbed clearly less water than those containing 'virgin' fibres. These differences were greatest at 40 % fibre content; composites containing RF absorbed approximately 0.5 time less water than WPC of 'virgin' fibres. The tendency for the volume change was the same but the differences were even greater. At 40 % fibre content, the absorbed water of the RF composites was only 20 % of that of the WPC containing 'virgin' fibres.

Table 6.4: Number of particles and its length (µm), referred to the length-based distribution (Q1) presented as 25% (x_{25}), 75% (x_{75}) quartiles and 50% (x_{50}) median prior to compounding; n=3, mean values (MV) ± standard deviation (SD).

	Number of particle	Particle length (µm)					
		x_{25}		x_{50}		x_{75}	
	MV	MV	SD	MV	SD	MV	SD
Virgin	245,161	474	±26	1188	±42	2016	±34
RF1	271,634	321	±25	997	±34	1941	±33
RF2	214,440	509	±10	1250	±18	2088	±18

Similar differnces in water uptake as without coupling agent were observed with MAPP (Table 6.5). At 60 % fibre content, the WPC containing RF took up less than 50 % water than the controls. The respective reduction in volume change amounted to more than 60 %.

Generally, using MAPP in the formulations decreased the water absorption at a maximum of 4.2 times. Incomplete encapsulation of wood fibres and probable occurrence of wood fibre agglomerates in the polymer matrix are additional influences on the water absorption properties (Butylina et al. 2011). All WPC specimens with 20 % fibre content slightly lost volume due to leaching of wood constituents. Addition of MAPP greatly decreased swelling in water up to 7.3 times.

This might be explained with a more hydrophobic surface of the RF and the better interaction with the PP matrix induced by the thermo-hydrolytic disintegration process and/or the remaining adhesives on the RFs’ surface (see above).

Table 6.5: Water uptake and volume change of the WPC with and without MAPP depending on the fibre type and the fibre content (HG); n=10, mean values (MV) ± standard deviation (SD).

		Water uptake (%)						Volume change (%)					
	MAPP	Virgin		RF1		RF2		Virgin		RF1		RF2	
HG	(%)	MV	SD	MV	SD	MV	SD	MV	SD	MV	SD	MV	SD
A	0	13.6	±0.3	10.3	±0.4	12.0	±0.1	12.1	±0.3	8.5	±0.5	10.0	±0.3
B	0	4.6	±0.2	2.1	±0.0	2.1	±0.0	2.9	±0.3	0.7	±0.2	0.9	±0.2
C	0	0.8	±0.0	0.5	±0.0	0.6	±0.0	-0.4	±0.2	-0.6	±0.8	-0.2	±0.2
D	3	7.8	±0.2	3.8	±0.0	2.8	±0.0	5.7	±1.3	2.5	±0.2	1.3	±0.3
E	3	1.8	±0.0	1.1	±0.0	1.2	±0.0	0.1	±0.4	-0.2	±0.2	-0.2	±0.3
F	3	0.6	±0.0	0.5	±0.0	0.5	±0.1	-1.2	±1.4	-0.2	±0.3	-0.4	±0.7

6.6 Conclusions

The performance of WPC made of RF was compared with that of WPC made of ‘virgin’ MDF fibres. RF and ‘virgin’ fibres in WPC formulations provided similar results, but composites containing RF exhibited improved mechanical and water-related properties than those containing ‘virgin’ fibres. With respect to mechanical properties, these differences were more prominent for flexural bending properties and unnotched impact bending, while differences in tensile strength were minor. These differences are attributed to enhanced compatibility between fibre surface and PP or to a higher flexibility of the RF. Using recovered MDF/HDF fibres for WPC production can ensure a more efficient utilisation of these wood resources.

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Chapter 7

The effect of fibreboard disintegration technique on wood polymer composites (WPC) produced with recovered wood particles

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Authorship

Table 7.0: Contribution of co-authors (as modified from Hunt 1991).

	Planning & Design	Executing & Data Collection	Data analysis & Interpretation	Literary input	Σ
F. Yağmur Bütün Buschalsky	25	25	20	25	**95**
Philipp Sauerbier	25	25	25	20	**95**
Holger Militz	0	0	5	0	**5**
Carsten Mai	5	0	20	10	**35**

F. Yağmur Bütün Buschalsky prepared the experimental design together with Philipp Sauerbier and Carsten Mai. Furthermore, she performed data collection together with Philipp Sauerbier. She analysed and interpreted the data and wrote the manuscript together with Philipp Sauerbier.

Philipp Sauerbier prepared the experimental design together with F. Yağmur Bütün Buschalsky and Carsten Mai. Furthermore, he performed data collection together with F. Yağmur Bütün Buschalsky. He analysed and interpreted the data and wrote the manuscript together with F. Yağmur Bütün Buschalsky.

Holger Militz supported the data interpretation.

Carsten Mai prepared the experimental design together with F. Yağmur Bütün Buschalsky and Philipp Sauerbier. He assisted the data interpretation and manuscript writing.

7.1 Abstract

The objective of this study is to evaluate and compare the mechanical and water-related properties of WPC containing two types of recovered wood particles. These particles were obtained by thermo-hydro-mechanical and sole mechanical (milling) disintegration and compared to virgin wood fibres. The respective composites were produced with and without maleic anhydride-grafted polypropylene (MAPP) as a coupling agent. The results clearly show that both types of particles, which derive from end-of-life MDF are a suitable resource to produce WPC. The physico-mechanical characteristics of the WPC containing particles from thermo-hydro-mechanical disintegration were statistically better than those containing mechanically produced particles.

7.2 Keywords

A. Wood fibres; A. Recycling; D. Mechanical testing; E. Autoclave

7.3 Introduction

Medium density fibreboards (MDF) are wood-based panels composed of roughly 80 % of ligno-cellulosic fibres, combined with a synthetic adhesive. These are joined together under heat and pressure to form panels predominantly in a dry process (Berglund and Rowell 2005; Irle and Barbu 2010). MDF find application mainly in the furniture industry due to their easy processability, acceptable weight-to-strength ratios and rather low costs. Thus, global MDF production together with that of high density fibreboards (HDF) has reached ca. 100 million m^3 in the year 2016 (FAO 2017). Currently, there is no commercially applied method to recycle waste MDF; therefore, MDF are burnt or landfilled (Irle et al. 2018) after end of life. The most advanced process is developed by BioComposite Centre, Bangor, UK together with the company MDF Recovery Ltd. which uses microwave technology for MDF disintegration. This process generates a new raw material source for the wood/natural fibre industry (BioComposite Centre 2015), but it is not industrially applied so far. A possible method to reuse discarded wooden products (including MDF) would be to disintegrate and transform them to new products (Wan et al. 2014). Various disintegration methods for wood-based panels (particleboard and MDF) have been developed throughout the years. These involve mechanical (Czarnecki et al. 2003), chemical (Irle et al. 2018), chemo-thermo-mechanical (Michanickl 1996) and thermo-hydrolytic (Roffael et al. 2010) disintegration to produce new particleboards and/or fibreboards. One of the possibilities of re-using waste MDF is processing them via a thermo-hydro-mechanical or mechanical disintegration process and then producing wood polymer composites (WPC) (Bütün et al. 2018).

WPC are a group of hybrid materials mainly consisting of renewable resources (Teuber et al. 2016c). They refer to composite products that contain wood (of any form) together with either thermoplastic or duroplastic polymers (Caulfield et al. 2005) and additives. The most commonly used systems for the compounding of WPC are counter-rotating and co-rotating twin-screw extruders (Schwendemann 2008). Production of the final product can be performed in a cold mould (injection moulding), between calenders (calendaring), or between two mould halves (compression moulding) (Stokke et al. 2014b). Mechanical properties of WPC depend on many factors, among the most important ones is the wood particle size (Gozdecki et al. 2011). The wood in the composite is generally classified in two major groups, fibrous and particulate (Wolcott and Englund 1999). Lignocellulosic materials can be used as fillers or reinforcements for thermoplastic polymers in industrial applications (Zimmermann and Zattera 2013). Small fibres with high aspect ratios (length to width ratio) are recommended for the use in WPC as they are distributed more homogeneously in the matrix than long fibres and provide a larger specific surface area, which improves the compatibility (Ashori 2008), thus, act as reinforcement. Wood particulates have dimensions that are approximately equal in all directions and can be of any shape (Wolcott and Englund 1999), they act mainly as fillers. However, the initial wood component morphology is not constant throughout WPC processing, but it is prone to changes due to shear forces and high temperatures, particularly during the compounding with twin screws in an extruder (Stark et al. 2004; Teuber et al. 2016a, b).

As the surface of wood is polar and the polypropylene used in this study is nonpolar, a coupling agent is often used to improve the compatibility between the polymer matrix and wood component (Gauthier et al. 1999; Caulfield et al. 2005). The use of maleic anhydride-grafted polypropylene (MAPP) has been the most common method to improve interfacial adhesion (Kim et al. 2007) in polyolefin-based WPCs due to its low cost and fair efficiency (Adhikary et al. 2008; Dányádi et al. 2010) as a coupling agent. There are various studies showing improved results for tensile strength, elongation at break and unnotched impact bending properties due to addition of MAPP (Oksman and Clemons 1998; Lu et al. 2000; Stark and Rowlands 2003; Gozdecki et al. 2015).

The objective of this study is to evaluate and compare the mechanical and physical properties of WPC containing two types of recovered wood particles. These particles were obtained by mechanical and thermo-hydro-mechanical disintegration and compared to results of using virgin wood fibres obtained from a previous study (Bütün et al. 2018). The respective composites were produced with and without MAPP as a coupling agent.

7.4 Material and Methods

7.4.1 Disintegration of the fibreboards

Two disintegration techniques were used for disintegrating surface laminated, UF-bonded (3.9 % N content) MDF from waste music speakers: thermo-hydrolytic and subsequently mechanical disintegration as well as mechanical disintegration. For thermo-hydrolytic disintegration, approx. 2 kg of MDF were first cut into pieces (50 x 50 mm in cross-section) and then immersed in 35 l water at 95 °C for 10 min in an autoclave (Zirbus Z3, Zirbus Technology GmbH, Bad Grund, Germany). During this process, the surface lamination detached from the MDF core and removed by hand. After drying to 5% moisture content at 70 °C, the thermo-hydrolytically recovered fibres were milled to wood flour in the grinding mill SM 2000 (Retsch, Haan, Germany) with 10 mesh size. The particles obtained were referred to as recovered particles from thermo-hydro-mechanically disintegrated fibreboards (TRF). For mechanical disintegration, the dry MDF were only ground by means of the aforementioned grinding mill and mesh size after the surface lamination has been removed manually. Recovered wood particles obtained by this technique are referred to as recovered particles from mechanically disintegrated fibreboards (MRF).

7.4.2 Composite preparation

The isotactic polypropylene (PP) Sabic 575P (Sabic, Riyad, Saudi Arabia) was used as polymer matrix; its density was 0.905 g cm^{-3}, and its melt flow index was 11 g (10 min)$^{-1}$ (230 °C 2.16 kg^{-1}). The maleic anhydride grafted polypropylene (MAPP) Licocene 7452 TP (Clariant, Muttenz, Switzerland) served as coupling agent; its density was 0.930 g cm^{-3} and grafting level was 7 %.

Compounding of the formulations containing TRF, MRF, PP and MAPP was processed by dry-blending of 40 wt% wood particles and 60 wt% PP; equal fibre content was compounded with 57 wt% PP at addition of 3 wt % MAPP (Table 7.1) using the co-rotating twin-screw extruder MICRO27GL/GG40D (Leistritz, Nürnberg, Germany) with two gravimetric feeders (DDW-MD5-

FW 40/5plus-50 ISC for wood componentand DDW-MD2-DSR 28-10 ISC for PP and MAPP components, Brabender Technology GmbH, Duisburg, Germany). The extruder was equipped with 9 heating zones, and the set temperatures from zone 1 to zone 9 were 180.0, 175.0, 175.0, 170.0, 170.0, 162.5, 152.5, 147.5 and 140.0°C, respectively. The average melting temperature was kept around 140 °C at the die, with an average screw speed of 121 rpm and pelletizer speed of 312.5 rpm.

WPC results obtained from virgin fibres (VF) containing 90 % Scots pine (*Pinus sylvestris*) and 10 % radiate pine (*Pinus radiata*) from a previous study of the authors (Bütün et al. 2018) served as a reference.

7.4.3 Injection moulding of the compounds

All compounded formulations were injection moulded using the injection moulding machine ALROUNDER 420 C (Arburg, Loßburg, Germany) at an average temperature of 174°C with pressure ranges between 160.4 and 204.4 kPa and average injection moulding cycle time of 44.0 s.

Table 7.1: WPC formulations (percent by weight).

WPC code	Wood particle type	Wood fibre content (%)	Polypropylene (PP) content (%)	Maleic anhydride grafted polypropylene (MAPP) content (%)
MRF 40 / 60	MRF	40	60	0
TRF 40 / 60	TRF	40	60	0
VF 40 / 60	VF	40	60	0
MRF 40 / 57 / 3	MRF	40	57	3
TRF 40 / 57 / 3	TRF	40	57	3
VF 40 / 57 / 3	VF	40	57	3

7.4.4 Dynamic image analysis of the wood particles

The particle size was determined prior to compounding using the dynamic image analysis system QICPIC combined with the dry dispersion unit RODOS (Sympatec GmbH, Clausthal-Zellerfeld, Germany). In the device, the wood particles are dispersed in an accelerated air jet and a high-speed camera takes pictures of the particle stream with a resolution of 10 µm and 20 µm. WINDOX software (Sympatec GmbH, Clausthal-Zellerfeld, Germany) was used to measure the particle size from the projected particle area of binary pictures (Teuber et al. 2016b). The particle size was characterized with respect to the median (x50) and quartiles (x25; smallest and x75; largest) of the length-based particle size distribution (Q1). The number of analysed particles per measurement varied between 561,387 and 1,428,774. Three replicate measurements were consecutively repeated for each of the wood particle type.

7.4.5 Characterisation of mechanical and physical properties

Rod-shaped specimens (80 x 10 x 4 mm) were used to determine flexural strength (ISO 178 2006), as well as unnotched impact bending (Charpy) (ISO 179 2006) and water absorption (EN 15534-1 2007)according to the standards. The water absorption of the specimens was gravimetrically assessed after 0, 1, 2, 4, 7, 21, and 28 days of immersion in water at 21±2 °C and related to the initial dry weight. Tensile strength (ISO 527-2 2003) was determined with dumbbell-shaped specimens (170 x 10 x 4 mm). Ten replicates were used for each test.

7.4.6 Determination of nitrogen content of the wood particles

The nitrogen content of the wood particles was determined by the Kjeldahl method for assessing the remaining UF binder on the wood particles after the two types of disintegration. Sulfuric acid (H_2SO_4) digestion of the samples was done using the device FoodALYT SBS 850 (Omnilab, Bremen, Germany). FoodALYT D 1000 (Omnilab, Bremen, Germany) was used for distillation of the acid digestion mixture to convert NH_4^+ to NH_3 by adding a small quantity of sodium hydroxide (NaOH), and condensation of the NH_3 gas in 2% boric acid (H_3BO_3) absorbing solution. Lastly, the solution was titrated manually to quantify the amount of ammonia in the boric acid solution by means of the device FoodALYT TS 10 (Omnilab, Bremen, Germany) and nitrogen percentage was calculated after three replicates for each wood particle type.

7.4.7 Statistical analysis

The obtained data were statistically analysed using Excel 2016 (Microsoft, Redmond, WA, USA). The Kolmogorov-Smirnov normality test ($\alpha = 0.05$) was applied to determine, if the results follow a normal distribution. A two sample unequal variance (heteroscedastic) t-test ($\alpha = 0.05$) was used to determine, if WPC properties with different wood particle types produced with and without MAPP differ significantly.

7.5 Results

Morphological analysis showed that the recovered particles (MRF, TRF) have roughly the same length distribution (Table 7.2). This applied to all three statistic intervals x_{25}, x_{50} and x_{75}. The virgin fibres, in contrast, were about 4.3 (x_{75}) to 5.5 (x_{25}, x_{50}) times longer than the recovered particles.

Table 7.2: Mean values of number of analysed particles and its length (µm), referred to the length-based particle distribution (Q1) presented as 25% (x_{25}), 75% (x_{75}) quartiles and 50% (x_{50}) median after grinding mill and prior to compounding; n=3, ± standard deviation.

Wood particle type	Number of analysed particles	Particle size (µm) x_{25}	 x_{50}	 x_{75}
MRF	561,387	83.0 (±0.8)	216.9 (±2.7)	466.3 (±5.1)
TRF	1,428,774	85.8 (±0.4)	213.4 (±2.5)	450.7 (±5.6)
VF	245,161	474.5 (±26)	1188.5 (±42)	2017 (±34)

The flexural strength (Figure 7.1) and the unnotched impact strength (Figure 7.2) of the MRF and TRF composites without coupling agent (40 / 60 formulations) did not differ significantly, while the tensile strength showed only minimal differences (Figure 7.3). The VF variant exhibited significantly lower values with respect to all strength properties than the composites containing MRF and TRF.

The addition of coupling agent (40 / 57 / 3 formulations) significantly increased the mechanical properties of all three variants (Figure 7.1-7.3). The values of all three mechanical properties for the TRF variant were slightly but significantly higher than for each of the MRF variants. WPC containing VF showed significantly lower flexural strength (Figure 7.1) and unnotched impact strength (Figure 7.3) than the MRF and TRF variants, while the tensile strength did not differ significantly (Figure 7.2).

The water absorption of variants containing recovered fibres (MRF, TRF) was clearly lower than that of WPC contain virgin fibres (Figure 7.4). The difference was significant from day 1 for formulations without coupling agent and from day 7 for formulations with coupling agent. The use of the coupling agent also led to a significant reduction in water absorption compared to respective variants without MAPP from day 1 onwards, regardless of the used particle. The water uptake of the TRF variants was significantly lower than that of the MRF variants. The difference occurs from day 4 for the variant without coupling agent (one exception on day 7) and from day 7 for the variant with coupling agent (one exception on day 14).

Thermo-hydro-mechanical disintegration reduced the nitrogen content of the TRF (2.0 %) compared to MRF (3.9 %), thus the remaining UF binder on the wood particles. The nitrogen content in the virgin fibres (VF) amounted to 0.1 %.

7.6 Discussion

The particle length distribution for both the MRF and the TRF was in a very similar range (Table 7.2). This is because both variants were milled with the same mesh size (10) in the grinding mill. The particle size is therefore not the cause for differences of the mechanical properties. Significant differences among the recovered particle variants occur only with the presence of MAPP; without the coupling agent the differences are statistically insignificant. It can therefore be assumed that the compatibility of wood and polypropylene without MAPP is not improved by residues of the UF adhesive. Instead, improved mechanical properties might be attributed to improved interaction of the coupling agent with the wood particle surface and the adhesive. The fibre flexibility might be enhanced through the hydrolytic treatment, which could result in higher strength properties. However, this assumption is not plausible due to the spherical particle structure and low aspect ratio (Table 7.2). Other reasons are more likely to explain the improvement in the mechanical properties due to previous hydrolysis of MDF. Firstly, the fibres are separated due to hydrolysis of the adhesive, which increases the surface of the separated TRF because of increased roughness, even if they have the same size after milling as the MRF. Roughness increase, however, is not detectable because of the resolution limitation of the optical measurement device used to determine the particle shape. Thus, there is generally more area for the MAPP to bind to wood and/ or to the UF binder. UF-based adhesives exhibit N-methylol or amino groups at their chain ends, which can form covalent or physical bonds with MAPP. More of these groups are available after hydrolytic disintegration even though the total adhesive content is lower, because in MRF the UF is partly inside the particles. In addition, cleavage of UF polymer chains gives rise to more amino and N-methylol end-groups. Simple grinding (MRF) exposes large surface areas, which are not covered with UF binder.

Another explanation is that the hydrolysis reverses hornification. Hornification is a term from pulp and paper research and refers to a partially reversible crosslinking and reduction of the volume of wood particles during a drying process (Minor 1994). Two mechanisms explain this behaviour. Crosslinking occurs within the fibre microstructure due to formation of interfibrillar hydrogen bonds between polysaccharide chains, which is (partially) irreversible. Another option involves chemical crosslinking.

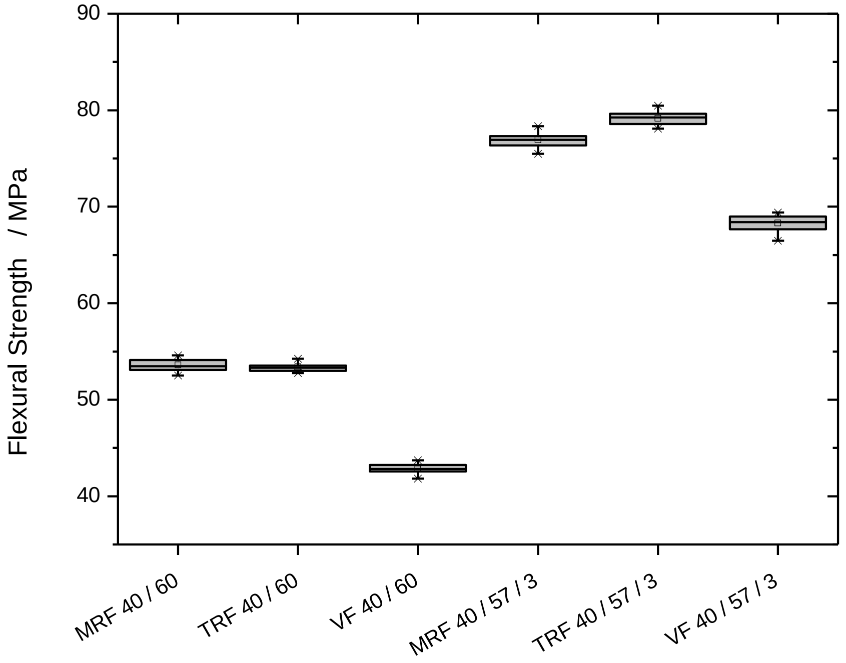

Figure 7.1: Flexural strength of WPC; n=10. The boxes of the plots are defined by the 25th and 75th percentiles.

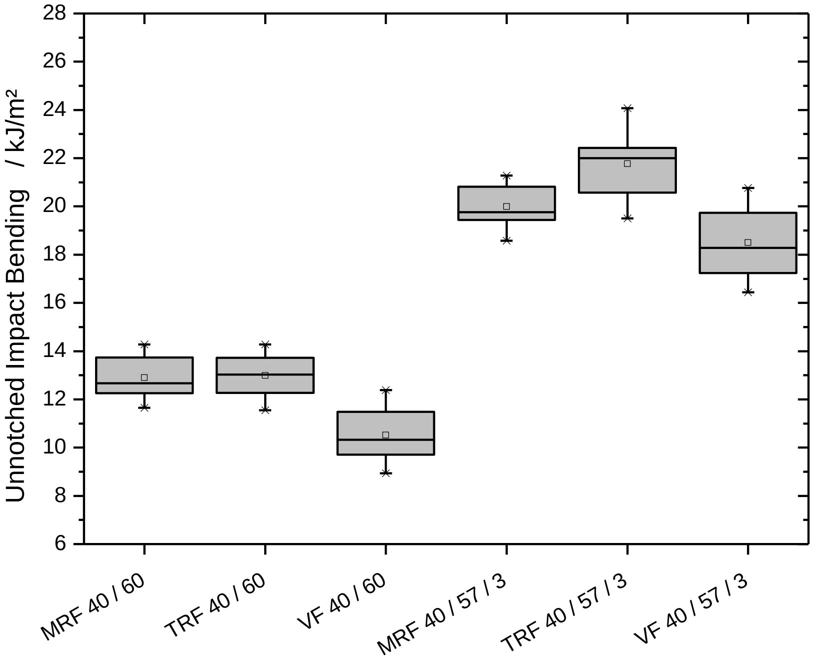

Figure 7.2: Unnotched impact bending strength of WPC; n=10. The boxes of the plots are defined by the 25th and 75th percentiles.

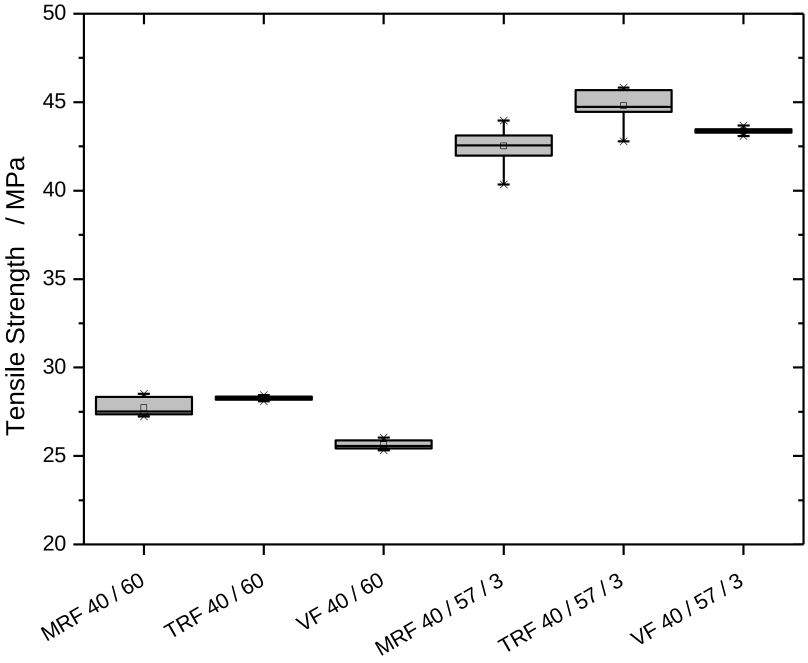

Figure 7.3: Tensile strength of WPC; n=10. The boxes of the plots are defined by the 25th and 75th percentiles.

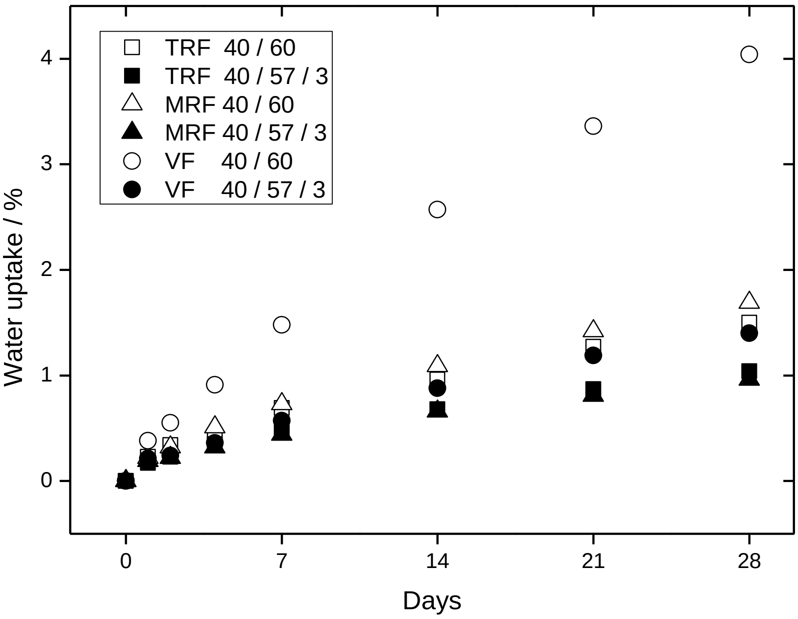

Figure 7.4: Water uptake of WPC; n=10. Two variants were analysed, without (40/60) and with (40/57/3) coupling agent.

In this case, lactones, e.g. with glucuronic side-chains (Fernandes Diniz et al. 2004) or acetal groups, e.g. with furfural, are formed. Irreversible hornification due to covalent cross-linking is induced by an acidic pH (Fernandes Diniz et al. 2004), high temperature and a drying process (removal of water molecules), which can cause the polysaccharides to rearrange (Kato and Cameron 1999). All conditions required for hornification are present during MDF production. It is therefore assumed that fibre hornification occurs during the MDF production process. The TRF process can cleave hydrogen bonds and partly covalent bonds. This increases the accessibility of MAPP to hydroxyl groups in TRF to form ester or hydrogen bonds.

The WPC produced with virgin fibres show less mechanical strength than those of the recovered particles. Even though the initial VF are much longer (Table 7.2), the mechanical properties are not better. The reasons can be found in the shortening of the VF during the extruding process, which equalizes size differences to a certain point. Additionally, as described earlier, the VF lack N-methylol or amino groups from the UF, therefore lacking these groups for bonding of the coupling agent, MAPP.

It should also be noticed that the physico-mechanical properties of the composites containing both types of recycled fibres and MAPP are within an expected range and are in accordance with previous studies (Sobczak et al. 2012).

The nitrogen content of MRF is the same as for the initial MDF (3.9 %). It can be considered as a measure of the initial binder content. This corresponds to a binder content of approximately 12 %, as the nitrogen content of UF binder is about 30 %. The thermo-hydro-mechanical disintegration reduced the nitrogen content of the TRF to 2.0 %, almost 50 % reduction. The binder has been transferred from the MDF particle’s surface into the disintegration water during the thermo-hydro-mechanical disintegration process. The nitrogen content of the virgin fibres (VF) was negligible (0.1 %). This indicates that major parts of binder material were still present in the particle mass (about 6 % related to the fibre mass), although significant hydrolysis and chain cleavage might have occurred.

7.7 Conclusions

The results of this study clearly show that end-of-life MDF are a suitable wood resource for WPC production. The mechanical properties achieved are in an acceptable range for the application of wood particle-filled polymers. While both disintegration techniques represent a suitable process to produce WPC from end-of-life MDF; the physico-mechanical characteristics are enhanced by the thermo-hydro-mechanical disintegration of the MDF in an autoclave before milling. However, there is only minor improvement due to the thermo-hydro-mechanical disintegration.

Additionally, the disintegration did reduce the nitrogen and therefore also the formaldehyde content of recovered wood material by extracting the adhesives. Which leads to a positive impact on emissions during service life.

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Chapter 8

Overview on results of the studies

8.1 Optimal thermo-hydrolytic disintegration conditions for disintegrating waste MDF

For disintegrating the waste MDF, thermo-hydrolytic disintegration process was introduced in Chapters 3, 4, 5, 6 and 7. It was found that waste MDF boards were softened and swelled after immersing the cut MDF pieces (5.0 x 5.0 cm^2 in cross-section) in pure water at 95°C for 15 to 30 min in an autoclave. The duration mostly depended on the amount, thickness and density of the immersed board and used resin type as well as amount of the water. The disintegration duration was increased while the listed parameters were increased and when the used resin type was MUF. Moreover, fibre bundles could still be found in the obtained RF, the further hammer-milling resulted in a roughly complete fibre separation. Thus, an effective disintegration was achieved. In contrast to aforementioned cut MDF pieces' cross-section dimensions, when the cross-section dimensions of the cut MDF pieces were reduced to 2.5 x 2.5 cm^2 as in Chapter 5, more amount of MDF pieces could be immersed, and therefore, disintegrated in lower amount of water.

8.2 Effect of the thermo-hydrolytic disintegration process on the obtained RF

There were prominent differences between the VF and RF in terms of chemical and geometrical properties which would be attributed to the high temperature during thermo-hydrolytic disintegration process, as well as the further hammer-milling process and the presence of the remaining resin after these processes.

8.2.1 Changes in the chemical properties

The pH values of the fibres (VF and RF) were measured after three different time spans (20 min., 4 h and 24 h), but the differences for each fibre type were so low that only the average values were reported. As a known fact, under acidic conditions in an aqueous environment, ammonia (NH_3) forms ammonium (NH_4) (Roffael and Hüster 2012; Wan et al. 2014). In Chapters 3 and 5, the slightly higher pH values of the RF were attributed to the formation of ammonia due to the breakdown of the UF-resin, which reacts to alkaline ammonium hydroxide (NH_4OH) in water (Roffael and Hüster 2012; Wan et al. 2014), thus reduced acidity of the RF.

Since the recovered fibres still contain cured UF resin, the formaldehyde emission of the RF were clearly higher than the VF. Furthermore, in Chapter 3, the second recycling process increased this difference even higher.

In contrast to the negligible low NC of the VF (0.07 %), the NC of the RF amounted to approximately 1.0 % after the first and 2.0 % after the second disintegration process in Chapter 3 and between 1.7 % and 3.3 % in Chapter 5. Using these NC values, the remaining resin content on the RF and the percentages of the initial resin content, which were between 33.5 % and 66.2 %, were calculated in both aforementioned chapters, respectively. These results upon those calculations revealed that the fibreboards have a relatively high tendency to be hydrolysed.

As the NC of UF resin is about 30 %, in Chapter 7, the NC of the MRF (3.9 %) was essential for finding the resin content of the waste MDF (12 %). In that study, the thermo-hydrolytic disintegration process reduced the NC of the TRF to 2.0 %, indicating that approximately half of the initial resin amount had been transferred from the recycled MDF pieces into the disintegration water and/or evaporated during the disintegration and drying processes.

8.2.2 Changes in the fibre geometry

In paper industry, it is well know that recycling leads to a shortening of the average fibre length, this phenomena corroborates also thermo-hydrolytic recycling of waste MDF (Grossmann 1996; Roffael et al. 2010). The presence of the elevated temperatures during thermo-hydrolytic disintegration process raises the possibility of shortening the fibres. Therefore, a greater amount of fine fibres are occurred and automatically reduce the average fibre length (Benthien et al. 2017).

Likewise, in Chapter 4, average fibre length and length-based distribution of VF measured by all three fibre analysers were always larger than the RF, while fibre widths of hammer-milled RF (except RF2-10, milled) were slightly wider (ca. 4.6 %) than those of VF.

In Chapter 6, even though they were all milled with the same mesh size (10) in the grinding mill, it was shown that the RF1 fibres were somewhat smaller than the VF, but RF2 fibre length was in the same range as the latter. Likewise, in Chapter 7, since both the MRF and the TRF were milled with the same mesh size (10) in the grinding mill, the particle size was therefore not the cause for the mechanical properties' differences between the produced WPC samples.

8.3 Chemical composition of the obtained DW

During the thermo-hydrolytic disintegration of waste MDF bonded with UF-resin, two hydrolysis reactions are occurring; the hydrolysis of wood components (mainly hemicellulose), which releases acetic acid and formic acid, thus, the pH is decreased (Packman 1960; Roffael and Hüster 2012), and the hydrolysis of the thermosetting UF-resin which is assumed to produce ammonia (NH_3) and carbon dioxide (CO_2), thus, the pH is increased (Wan et al. 2014). Moreover, as the cured resins with high U:F molar ratio contain a small amount of still unreacted urea (Hameed et al. 2006), free urea, after its transformation to ammonia, also forms ammonium ions, especially in presence of acids generated either from wood or from the used hardener (Roffael et al. 2018).

In Chapter 3, the decreased pH of the DW can be attributed to the degradation of the wood components, especially hemicelluloses, during the thermo-hydrolytic disintegration process. Due to the amorphous structure and presence of the acetyl groups, hemicelluloses are less thermal stable than cellulose and lignin (Hill 2006). Thus, during the thermo-hydrolytic disintegration process hemicelluloses have the highest degree of degradation compared to other wood macromolecules, which results in the cleavage of the acetyl groups and glyosidic bonds. Moreover, the analysed significant levels of reducing sugar and equivalents in the DW after the first thermo-hydrolytic disintegration process (also as in Chapter 5) support this attribution.

Furthermore, the analysed excessively low levels of reducing sugar and equivalents in the DW after the second thermo-hydrolytic disintegration process in Chapter 3 indicate that most of the extractable sugars were removed already after the first disintegration process.

In Chapter 3 and 5, higher NC of DW samples is an indication of containing more dissolved/hydrolysed UF resin in the sample. Consistently, the higher formaldehyde content in the disintegration water indicates the higher degree of hydrolysis of the resin (Lubis and Park 2018).

8.4 Utilisation of RF for manufacturing MDF panels

Regarding the physical, mechanical, chemical and formaldehyde related properties of the manufactured recycled MDF panels, some results showed inconsistent trends when different board thicknesses were compared.

Results from previous studies reported that up to 30 % of RF can be a partial substitute of TMP from wood in manufacturing UF-resin bonded recycled MDF without deteriorating the physico-mechanical properties of the panels (Roffael et al. 2010).

Due to their easy accessibility, hemicelluloses are the main compound for hydrogen bonding on wood surfaces (River et al. 1991; Frihart 2005; Popovic et al. 2015). Moezzipour et al. (2018) attributed the higher TS of recycled MDF panels to the removed lignin on the surface of RF, thus increased crushing of them. However, in Chapter 3, only TS values of 16mm recycled MDF panels were greater than the reference panels. The presence of N from the remaining UF-resin on the RF should have caused poor adhesion in the recycled MDF panels. Furthermore, the higher IB values of these panels can be attributed to their higher densities.

Esterification reactions have been occurred in RF which will result in inferior mechanical properties of the fibres and recycled MDF panels manufactured with them due to their effect on the polymerization of the UF-resin (Moezzipour et al. 2017). Roffael et al. 2016 and Behn et al. 2018 recommended using a small amount of pMDI in addition to UF-resin during the manufacturing of the recycled MDF panels for reducing this effect.

Urea, ammonia and oligomeric decay products of UF resin can react with formaldehyde and act as formaldehyde scavengers. In Chapter 3, the formaldehyde emission determined by perforator method of the recycled panels was lower than that of virgin panels throughout the different generations of the study. However, in Chapter 5, the formaldehyde emissions of the recycled MDF panels were significantly higher than the original MDF panels. A possible explanation was that the original MDF panels which were used in Chapter 5 were manufactured industrially and limitations of the laboratory-scale production of the recycled MDF panels could cause some inconsistent physico-mechanical and chemical results when panels were compared.

Since the fibre production stage has the highest environmental impact due to high energy use, chemical additive production and transportation burdens, reducing the total requirement for virgin fibres should reduce environmental impacts (Mitchell and Stevens 2009). The findings of this thesis when it comes to utilising RF for manufacturing new MDF panels can contribute to this phenomena.

8.5 Utilisation of RF for manufacturing WPC

In Chapter 6, the results indicated that WPC containing RF exhibited higher tensile and flexural strengths as well as impact bending compared to that containing VF, while the moduli of the composites containing RF were the same as those containing VF. The slightly higher strength properties of WPC containing RF were attributed to the better compatibility of the RF surface with the polypropylene matrix, due to the remaining UF-binder on the RF.

Correspondingly, in Chapter 7, WPC containing RF obtained after thermo-hydrolytic disintegration of waste MDF exhibited higher mechanical properties compared to those containing VF and RF obtained after solely mechanical disintegration of waste MDF.

Taken together, these results suggest that the thermo-hydrolytic disintegration process didn’t negatively affect the mechanical properties of the WPC compared those containing VF, even though, the initial fibre length of the VF was longer. On the other hand, the water absorption of the WPC containing RF after thermo-hydrolytic disintegration of waste MDF was clearly lower than that of WPC containing VF and RF obtained after solely mechanical disintegration of waste MDF. Furthermore, as RF material contains N-methylol and/or amino groups from the hydrolysed UF-resin, the bonding between the coupling agent and the RF might have been improved.

8.6 Short summary of the results

This study has gone some way towards enhancing the understanding of waste MDF recycling and the possible utilisation areas for obtained RF after the thermo-hydrolytic disintegration of them by answering the initially stated research questions. For obtaining RF, a thermo-hydrolytic disintegration technique of waste MDF was successfully introduced in an autoclave. This technique was performed at significantly lower temperature (at 95 °C) using solely water and took clearly shorter duration than in previous studies. As a result of the thermo-hydrolytic process, recovered fibres and disintegration water were obtained.

Obtained RF were found to be shortened compared to VF due to the disintegration conditions and to have altered chemical properties, resulting in higher pH and formaldehyde emissions due to the remaining resin.

Moreover, RF were further utilized for manufacturing either new MDF panels or WPC. When utilized for manufacturing new MDF, even the utilisation of 100 % RF did not cause significantly lower strength properties than those of virgin panels. However, introducing the screw press process for drying the RF after the disintegration caused decreased strength properties compared to the original MDF panels containing 100 % VF. Although, mixing screw-pressed and air-dried RF with VF improved these properties, the strength of the panels containing solely VF were not achieved. Furthermore, WPC formulations containing RF exhibited caused improved mechanical and water-related properties, even though they provided similar physico-mechanical results to those containing VF.

The study has also shown that obtained DW exhibited higher pH values, N as well as formaldehyde contents while containing significant levels of reducing sugar and equivalents when compared to demineralized water.

The findings of this dissertation not only make several scientific contributions to the current literature, but also suggest utilizing RF obtained after the thermo-hydrolytic disintegration of waste MDF for manufacturing new MDF and WPC to ensure a more efficient utilization of these wood resources. Further research might be carried out to explore how different mixing proportions of RF and VF effect the strength properties of these possible utilisation areas, especially new MDF.

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CCA	copper chrome arsenic
CH_2O	formaldehyde
CH_3OH	carbamic acid
CH_4N_2O	urea
CMOS	complementary metal-oxide semiconductor
CNC	cellulose nanocrystals
CO_2	carbon dioxide
CTMP	chemo-thermo-mechanical pulp
DIA	digital image analysis
DNA	dinitrosalicylic acid
dpi	dot per inch
DW	disintegration water
EC	European Commission
EU	European Union
FB	fibreboard
FTIR	Fourier-transform infrared spectroscopy
GHz	gigahertz
GL	geodesic length, fibre length
GW	fibre width
H_2SO_4	sulphuric acid
H_3BO_3	boric acid
HDF	high density fibreboard
HDPE	high-density polyethylene

HPLC	high-performance liquid chromatography
HRF	hammer-mill recovered fibres
IB	internal bond strength
LVL	laminated veneer lumber
MAPE	maleic anhydride-grafted-polyethylene
MAPP	maleic anhydride polypropylene
MC	moisture content
MDF	medium density fibreboard
MHz	Megahertz
MOE	modulus of elasticity
MOR	modulus of rupture
MRF	particles from mechanically disintegrated fibreboards
MUF/mUF	melamine-urea-formaldehyde
MV	mean value
n/a	not applicable
n. a.	not analysed
n. c.	not calculated
N	nitrogen
NaOH	sodium hydroxide
NC	nitrogen content
NH_2	amine
NH_3	ammonia
NH_4	ammonium

NaOH	sodium hydroxide
Na_2S	sodium sulphide
NH_4OH	ammonium hydroxide
$(NH_4)_2SO_4$	ammonium sulphate
NO_x	nitrogen oxides
o.d.	oven dried
OSB	oriented strand board
PB	particleboard
PE	polyethylene
PF	phenol formaldehyde
pMDI	polymeric diphenylmethane diisocyanate
PP	polypropylene
PW	plywood
RF	recovered fibres
RRF	refiner recovered fibres
SD	standard deviation
TH	thermo-hydrolytic
THE	thermo-hydro-electrical
TMP	thermo-mechanical pulp
TRF	particles from thermo-hydro-mechanically disintegrated fibreboards
TS	thickness swelling
TSE	twin-screw extruder
UF	urea-formaldehyde

VF	virgin fibre
WBP	wood-based panels
WPC	wood polymer composites
WU	water uptake
µm	micrometre

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