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

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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, the focus was 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} was subjected to thermo-hydrolytic disintegration in an autoclave using only water at 95°C for 20–30 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.

1 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 has reached 98.6 million m^3 in 2018 (FAO 2019). 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

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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 80 s, 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.

2 Material and methods

2.1 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 hammer mill (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 $((\text{NH}_4)_2\text{SO}_4)$ 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 hammer mill with the same mesh size. Subsequently, fibre mats were manually formed using a $450 \times 450 \text{ mm}^2$ 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 1). After a day of cooling down and subsequent trimming the edges of the panels to $400 \times 400 \text{ mm}^2$, 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 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

2.2 Thermo-hydrolytic disintegration of MDF panels

For each disintegration run, pieces cut from first or subsequent generation MDF with $50 \times 50 \text{ mm}^2$ 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 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 L of the disintegration water.

2.3 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 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.

2.4 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 taken 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).

Table 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, 10 mm	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

2.5 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 $50 \times 50 \times$ 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 $250 \times 50 \times 10 \text{ mm}^3$ and $370 \times 50 \times 16 \text{ mm}^3$ 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 previously described (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 ($<0.1\%$).

2.6 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 previously described (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).

2.7 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 Results and discussion

3.1 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). While the pH value of VF amounted to 4.0, those of RF were on 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

Table 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	10 mm	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
	16 mm	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	10 mm	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
	16 mm	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”

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). 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 Cowlings 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 negligibly low, the NC of the RF amounted to approximately 1%

after the first and 2% after the second disintegration process (Table 3). As the NC of the virgin MDF panels was approximately 3% (Table 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).

3.2 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 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

Table 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 emission—Perforator method (mg/100 g o.d. board)	Formaldehyde emission—Gas analyser method (mg/m ² h)	Formaldehyde emission—Flask method (mg/kg o.d. board) (3 h)	Formaldehyde emission—Flask method (mg/kg o.d. board) (24 h)	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
	16 mm	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

Table 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) (%) 24 h	Thickness swelling (TS) (%) 24 h
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}

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, 2010a; Lykidis and Grigoriou 2008; Bütün et al. 2018, 2019; 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 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 in 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 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 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 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. Moezzi-pour 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 generation 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. 2010b; 2016; Behn 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 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 emission 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 emission 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. 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 in 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. 2010a), 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 remaining 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 physico-mechanical 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.

3.3 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 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.

Table 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”

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 (pulp- ing) 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 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) 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.

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 L of water, on average, approximately 65% and 50% of the initial UF was found in the DW samples from the 1st and the 2nd disintegration process, 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.

4 Conclusion

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 (pulp- ing) and/or thermo-hydrolytic recycling.

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Declarations

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