

*SHORT NOTES***APPLICATION OF MINERAL FILLER IN MEDIUM DENSITY FIBERBOARD (MDF) AND ITS EFFECT ON MATERIAL PROPERTIES AS A FUNCTION OF PARTICLE SIZE**

TOMASZ OZYHAR
OMYA INTERNATIONAL AG
SWITZERLAND

(RECEIVED DECEMBER 2020)

ABSTRACT

The addition of inorganic filler material in medium density fiberboard (MDF) and the effect on material properties as a function of particle size was examined. Medium density fiberboard was manufactured in a laboratory scale environment to a target raw density of 750 kgm^{-3} . Wood fibers were replaced by using calcium carbonate at 3 and 10 wt.% using fillers with weighted median particle sizes of $d_{50} = 2.0 \text{ }\mu\text{m}$ and $d_{50} = 30 \text{ }\mu\text{m}$, respectively. Urea formaldehyde resin was used as binder in all MDF. The influence of filler addition on the modulus of elasticity, bending and tensile strength, dimensional stability and liquid permeability was investigated. The results demonstrate the effect of filler content and its dependence on particle size. The addition of filler with $d_{50} = 30 \text{ }\mu\text{m}$ does not have any influence on material properties up to a filler content of 10 wt.%. Using the finer filler with $d_{50} = 2.0 \text{ }\mu\text{m}$ at 10 wt.% filler, the quantity significantly increases the water adsorption and swelling behavior and reduces the strength properties of the MDF.

KEYWORDS: Dimensional stability, liquid permeability, mechanical properties, medium density fiberboard (MDF), mineral filler, particle size, urea formaldehyde.

INTRODUCTION

Medium density fiberboard (MDF) usually consists of wood as the main raw material, complemented by typically 3 to 15% of a thermoset adhesive (solid resin based on dry mass of wood) and, depending on the final composition, selected additional additives such as hardener, water repellents, dyes and inks, etc. (Rowell 2012). In contrast, inorganic minerals are rarely used in this kind of application and have received little attention. Limited studies on the use of inorganic minerals in engineered wood base panels have focused predominantly on improving the fire resistance properties by incorporating inorganic minerals such as aluminum trihydrate

(ATH) (Hashim et al. 2005, Redwan et al. 2015), nanoclay (Zahedsheijani et al. 2012) or expanded vermiculite (Wang et al. 2016). At the same time, the application of mineral fillers, especially calcium carbonate as a cost competitive material to replace cellulose fiber, with the goal to reduce manufacturing cost, is well known in the paper industry (Hubby and Gill 2016). Considering the obvious similarities in the structure of MDF and paper, it is somehow striking that a similar concept, i.e. the potential utilization of calcium carbonate as a filler to partially replace wood fibers has not been investigated in detail until now. Only recently, the use of a mineral additives as a partial substitute for wood material in MDF and particle board was suggested in Ozyhar 2020 and Ozyhar et. al 2020. While the authors have investigated the influence of calcium carbonate addition on basic material properties in relation to addition quantity, the effect from particle size on properties including the mechanical strength or water absorption, remains unanswered. However, particle size of the mineral filler is a known factor which have been shown to be critical to strength development in paper (Bown 1998, Hubby and Gill 2016).

The following study was aimed to create a basic understanding regarding the potential for use of an inorganic filler as a partial replacement for wood fibers in MDF. Specifically, the main goal was to evaluate the influence of calcium carbonate addition on the material properties of MDF as a function of particle size.

MATERIAL AND METHODS

Materials

Fresh wood fibers, obtained from pine wood chips, were used for MDF manufacturing. These were broken down in a refiner at 9 bar for 3 to 4 min. The sieve analysis of the fiber material measured by fractioning through sieve analysis in an air jet sieve Alpine e200 LS of HOSOKAWA ALPINE AG, Germany, revealed that 97% were smaller than 2.0 mm, 86% smaller than 1.0 mm and 33% smaller than 0.2 mm, respectively. Two ground calcium carbonate (GCC) fillers with differing particle sizes were used. A finer filler with weighted median particle size $d_{50} = 2.0 \mu\text{m}$ and surface area of 2.7 m^2 (FGCC) and a coarser filler with weighted median particle size $d_{50} = 30.0 \mu\text{m}$ and surface area of 1.2 m^2 (CGCC). A formaldehyde resin, Kaurit 350 supplied by BASF AG and a hydrophobing agent, Hydrowax 138 supplied by SASOL GmbH were used in this study.

Methods

Four different MDF variants were manufactured by replacing the respective amount of wood fibers by weight using calcium carbonate fillers with filler quantities of 3 and 10 wt.% and the different particle sizes, fine (FGCC) and coarse (CGCC), respectively. MDF with a target density of 750 kg m^{-3} was manufactured in a laboratory scale environment by mixing the fibers in a paddle mixer with 10 wt.% urea formaldehyde resin and 0.5 wt.% hydrophobing agent (based on wood fibers). Calcium carbonate was added in powder form to the paddle mixer and mixed with the fiber after the urea formaldehyde resin had been applied to fiber. MDF with target density of 750 kg m^{-3} was manufactured in laboratory scale environment by mixing the fibers in

a paddle mixer with 10 wt.% urea formaldehyde resin and 0.5 wt.% hydrophobing agent (based on wood fibers). The fiber/filler mix was then pressed into a 460 x 440 mm² solid board of 17.5 mm thickness at a temperature of 220°C using a press time factor of 12 s·mm⁻¹. The boards were then sanded to dimensions of 400 x 380 mm² and final thickness of 16 mm.

Internal bond strength (IB), bending strength (BS) and modulus of elasticity in bending (MOE), water absorption (WA) and thickness swelling (TS) behavior after 24h and the raw density were determined on the samples acclimatized at 20°C and relative humidity of 65% following the procedures described in DIN EN 310 (1993), DIN EN 317 (1993), DIN EN 319 (1993) and DIN EN 323 (1993). In addition, liquid permeability was determined on 4 samples for each variant following the test method described in Ozyhar et. al (2020).

RESULTS AND DISCUSSION

To ensure homogeneity between the filler addition and the fibers, resin was added to the fibers prior to adding the filler to guarantee the adherence of the filler to the fibers. The filler distribution in the fiber matrix was investigated visually using SEM images, with specific interest on any potential agglomeration of particles that could influence locally the material strength and other properties. The uniform distribution of calcium carbonate filler in the wood fiber matrix, as shown in Fig. 1, eliminates the potential influence of uneven filler distribution or agglomeration on the material properties.

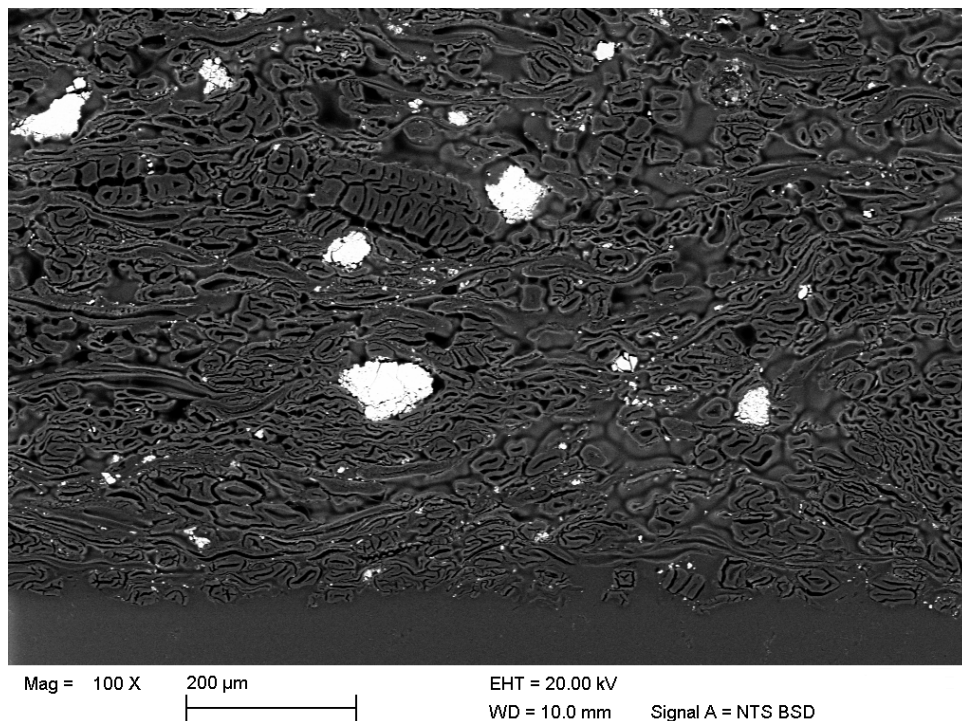


Fig. 1: Scanning electron micrograph (SEM) of cross section of medium density fiberboard (MDF) with addition of 10 wt.% of coarse calcium carbonate (CGCC) as filler.

Filler effect on mechanical properties

The influence of calcium carbonate addition as a filler material in MDF on the material properties measured is summarized in Tab. 1. The results demonstrate that filler content has an effect on the material properties and there is a dependence on the particle size.

Tab. 1: Material properties of medium density fiberboard (MDF) with 3 and 10 wt.% of calcium carbonate as filler.

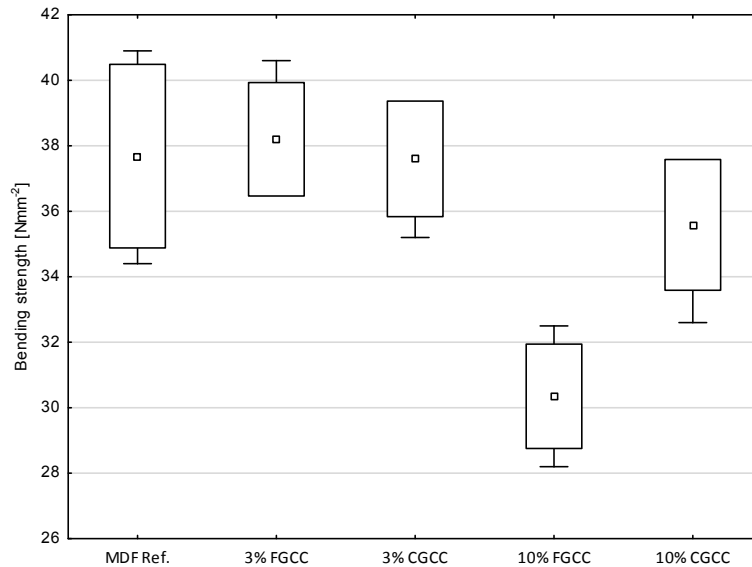
Material property		Sample nomenclature				
		0% filler	3% filler		10% filler	
		Reference MDF	FGCC	CGCC	FGCC	CGCC
Raw density ($\text{kg}\cdot\text{m}^{-3}$)	\bar{x}	770	767	778	763	761
	(CV%)	(2.2)	(1.8)	(1.8)	(2.3)	(3.0)
	n (-)	10	10	10	10	10
BS (Nmm^{-2})	\bar{x}	3.77	30.8	30.7	30.4	35.6
	(CV%)	(7.1)	(4.9)	(3.6)	(5.0)	(5.3)
	n (-)	6	6	6	6	6
IB (Nmm^{-2})	\bar{x}	1.66	1.59	1.62	1.23	1.67
	(CV%)	(4.2)	(5.3)	(3.5)	(5.5)	(3.0)
	n (-)	10	10	10	10	10
MOE (Nmm^{-2})	\bar{x}	3310	3327	3253	2902	3030
	(CV%)	(5.6)	(3.7)	(3.4)	(4.6)	(4.9)
	n (-)	6	6	6	6	6
WA (%)	\bar{x}	20.6	24.0	22.4	31.5	24.8
	(CV%)	(11.4)	(11.4)	(5.0)	(7.3)	(7.3)
	n (-)	10	10	10	10	10
TS (%)	\bar{x}	6.1	6.4	6.1	8.3	6.5
	(CV%)	(9.1)	(5.5)	(2.8)	(4.4)	(3.7)
	n (-)	10	10	10	10	10
Permeability ($10^{-13}\cdot\text{m}^2$)	\bar{x}	2.2	2.0	2.0	3.5	3.3
	CV(%)	(15.4)	(10.3)	(30.4)	(28.0)	(13.4)
	n (-)	4	4	4	4	4

Note: \bar{x} - arithmetic mean, CV - coefficient of variation, n - number of specimens, FGCC - fine ground calcium carbonate, CGCC - coarse ground calcium carbonate, BS - bending strength, IB - internal bond strength, MOE - modulus of elasticity, WA - water adsorption, TS - thickness swelling.

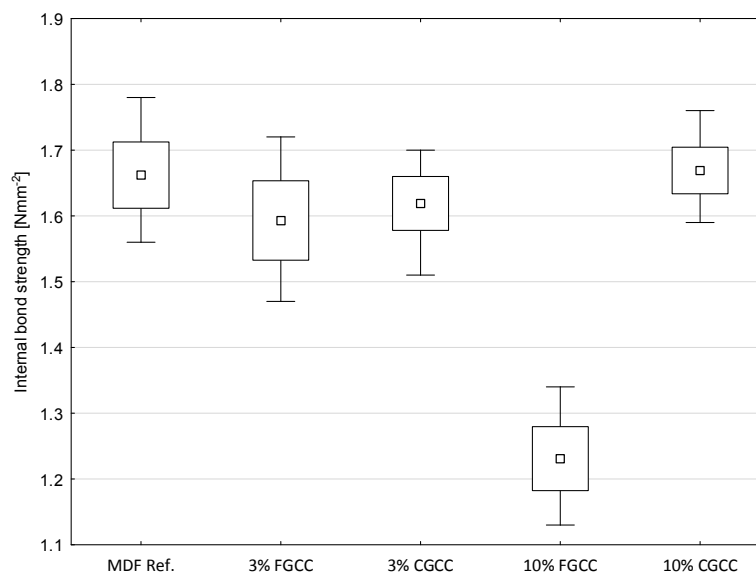
The relationship becomes most evident for the mechanical properties bending and internal bond strength (Figs. 2a,b). Addition of CGCC filler does not have a significant effect on the strength properties up to a filler content of 10 wt.%. However, strength properties are affected significantly at loading levels of 10 wt.% when using FGCC indicating that effect from filler content depends on particle size.

It is suggested that the mechanical strength behavior seen is directly related to the surface area of the particles and the adhesive bond formation between wood fibers, directly related to

strength. The FGCC has a surface area more than double that of the CGCC, $2.7 \text{ m}^2\text{g}^{-1}$ and $1.2 \text{ m}^2\text{g}^{-1}$, respectively.



a)



b)

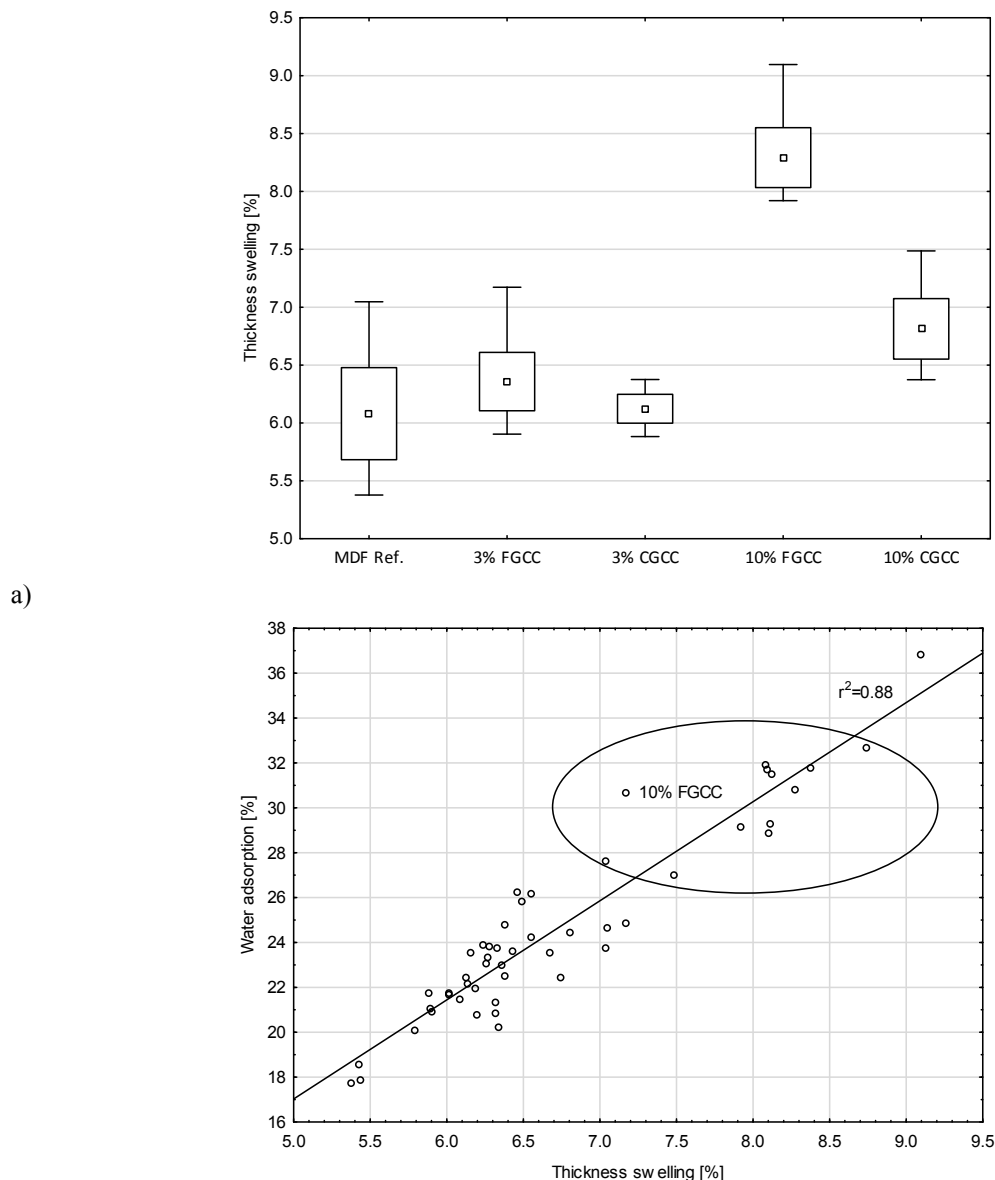
Fig. 2: a) Bending strength and b) internal bond strength as function filler content and filler particle size. Whiskers show minimum and maximum values, square in box displays arithmetic mean value, box boundaries display mean \pm 0.95 conf. interval. MDF. Ref. (medium density fiberboard without filler addition); FGCC and CGCC refer to fine and coarse ground calcium carbonate, resp.

The FGCC particles are believed to affect the adhesive bond formation to a much higher extent firstly by reducing the surface available on the wood fibers for bond formation and mechanically disturbing the wood fibers themselves. Secondly, the FGCC particles adsorb and “consume” more resin per unit mass with increasing surface area thus reducing the available resin for fiber bond formation.

The high surface area, in combination with an expected negative effect on UF curing associated with alkaline surface pH of calcium carbonate is believed to be the main reason for decline in strength seen with decreasing particle size. The negative effect of increased pH on curing kinetics of UF and bond formation have been demonstrated previously (Johns and Niazi 1980, Pizzi 2003).

Filler effect on physical properties

The effect of filler addition on the dimensional stability of MDF is clearly demonstrated by the thickness swelling measurement (Fig. 3a).



b) Relationship between water adsorption and swelling behavior for caused by calcium carbonate addition.

MDF. Ref. (medium density fiberboard without filler addition); FGCC and CGCC refer to fine and coarse ground calcium carbonate, resp.

Only at the higher loading level is there any change in TS. The small increase in thickness measured at 10 wt.% for the CGCC is not statistically significant, therefore, no effect on thickness swelling is concluded for this sample. Fine GCC filler, on the other hand, when applied at 10 wt.%, increases the corresponding TS significantly. The TS is, therefore, effected by the filler content percentage and depends on the particle size. Fig. 3b shows the water adsorption following an almost linear trend in relation to the thickness swelling. The results show that by adding FGCC filler to MDF the water uptake increases and correlates closely with increasing thickness swelling.

The findings are contradictory to what expected by replacing 10% of hydrophobic wood fibers with inorganic material, i.e. decreased swelling due to less hydrophobic material available. This is likely due to the increased liquid permeability of the MDF at a filler addition level of 10 wt.% (Fig. 4).

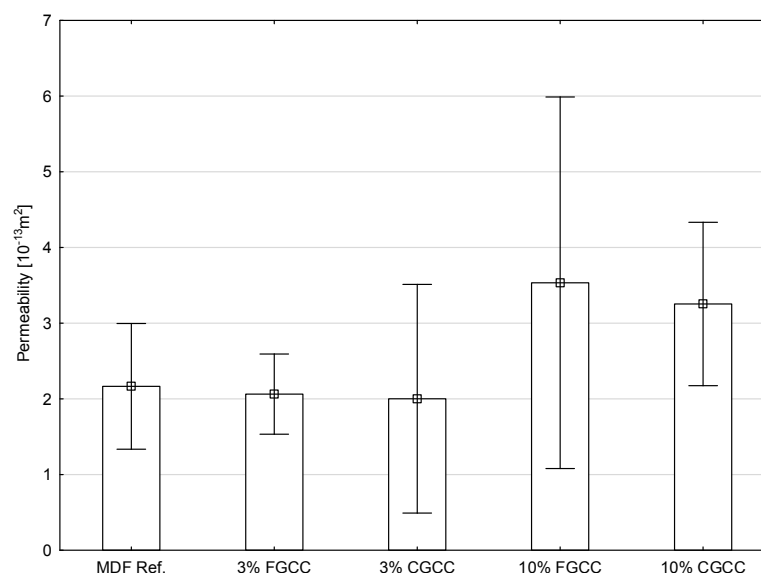


Fig. 4: Permeability of medium density fiberboards with 3 and 10 wt.% calcium carbonate addition as filler. Whiskers show conf. interval at 0.95 level, square in box displays arithmetic mean value. MDF. Ref. (medium density fiberboard without filler addition); FGCC and CGCC refer to fine and coarse ground calcium carbonate, resp.

An increased water uptake was expected for MDF panels at 10 wt.% filler content based on liquid permeability values measured using hexadecane, a method known to determine liquid permeability in paper materials (Ridgway and Gane 2003). The higher permeability values of the MDF with the higher filler addition when compared with control panel, can be explained by the packing density of the fiber-filler furnish and the much higher density of the filler. Having a higher density, calcium carbonate occupying less volume increases the theoretical porosity and consequently the permeability at constant volume and mass of panel.

CONCLUSIONS

The results of this study prove that the addition of calcium carbonate (GCC) as a filler in MDF has an effect on the material properties studied here. Filler quantity and particle size play an important role. The results demonstrate that the filler with weighted median particle size (d_{50}) of 30 μm of up to 10 wt.% can be added to MDF without effecting the material properties. However, addition of GCC filler consisting of small particles with particle size $d_{50} \leq 2 \mu\text{m}$ has a significant effect on all material properties leading to a decrease in the mechanical properties and an increase in the water adsorption and swelling thickness, believed to be caused by increased permeability.

ACKNOWLEDGMENTS

The author would like to express his deepest gratitude to Mr. Weber and Mr. Direske from Institut für Holztechnologie Dresden (IHD) for supervising of manufacturing of MDF samples and determination of material properties. Special thanks go to Dr. Ridgway for performing the permeability measurements and reviewing the manuscript.

REFERENCES

1. Bown, R., 1998: Particle size, shape and structure of paper fillers and their effect on paper properties. *Paper Technology* (39): 44-48.
2. DIN EN 310, 1993: Wood-based panels. Determination of modulus of elasticity in bending and of bending strength.
3. DIN EN 317, 1993: Particleboards and fiberboards. Determination of swelling in thickness after immersion in water .
4. DIN EN 319, 1993: Particleboards and fiberboards. Determination of tensile strength perpendicular to the plane of the board.
5. DIN EN 323, 1993: Wood-based panels. Determination of density.
6. Hashim, R., How, L.S., Kumar, R.N., Sulaiman, O., 2005: Some of the properties of flame retardant medium density fiberboard made from rubberwood and recycled containers containing aluminum trihydroxide. *Bioresources Technology* 96: 1826-31.
7. Hubbe, M.A., Gill, R.A., 2016: Fillers for papermaking: A review of their properties, usage practices, and their mechanistic role. *BioResources* 11: 2886-2963.
8. Johns, W.E., Niazi, K.A., 1980: Effect of pH and buffering capacity of wood on the relation time of urea formaldehyde resin. *Wood and Fiber Science* 12: 255-263.
9. Ozyhar, T., 2020: Application of mineral filler in surface layer of three-layer particle board and its effect on material properties as a function of filler content. *International Wood Products Journal* 11(3): 109-114.

10. Ozyhar, T., Depnering, T., Ridgway, C., Welker, M., Schoelkopf, J., Mayer, I., Thoemen, H., 2020: Utilization of inorganic mineral filler material as partial replacement for wood fiber in medium density fiberboard (MDF) and its effect on material properties. *European Journal of Wood and Wood Products* 78(1): 75–84.
11. Redwan, A.M., Badri, K.H., Tarawneh, M., 2015: The effect of aluminium hydroxide (ATH) on the mechanical properties and fire resistivity of palm-based fibreboard prepared by pre-polymerization method. *Advanced Materials Research* 1087: 287-292.
12. Pizzi, A., 2003: Urea–formaldehyde adhesives. In: *Handbook of adhesive technology*, second edition, revised and expanded. Eds. Pizzi A., Mittal, K.L. Pp 613-631, CRC Press, New York, NY.
13. Ridgway, C.J., Gane, P.A.C., 2003: Bulk density measurement and coating porosity calculation for coated paper samples. *Nordic Pulp & Paper Research Journal* 18: 24–31.
14. Rowell, R.M., 2012: *Handbook of wood chemistry and wood composites*. 2nd Edition. Pp 321-413, CRC Press, Boca Raton.
15. Wang, J., Wang, F., Gao, Z., Zheng, M., Sun, J., 2016: Flame retardant medium-density fiberboard with expanded vermiculite. *BioResources*. 11: 6940-6947.

*TOMASZ OZYHAR
OMYA INTERNATIONAL AG
RESEARCH AND DEVELOPMENT SERVICES
FROSHACKERSTRASSE 6
EGERKINGEN
SWITZERLAND

*Corresponding author: tomasz.ozyhar@omya.com