WASTE AGGLOMERATED WOOD MATERIALS AS A SECONDARY RAW MATERIAL FOR CHIPBOARDS AND FIBREBOARDS PART I. PREPARATION AND CHARACTERIZATION OF WOOD CHIPS IN TERMS OF THEIR REUSE

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ABSTRACT

The article describes a method of preparing particles from waste particle boards (chipboards) and oriented strand boards (OSBs). Their reuse is the main target of recycling. Method of their destruction was determined in this work. Agglomerated materials disintegrated after an initial destruction were further processed under specified conditions with regard to the material humidity, type of materials, contained adhesives and given characteristics of final particles - wood chips. Wood particles obtained were characterized by a fractional composition and amount of residual formaldehyde as an important parameter for the reuse of waste materials in production of furniture boards. New chipboards and pulp for production of middle density fiberboards (MDFs) will be provided from such defined particles.

KEY WORDS: Recycling of wood materials, waste chipboard, OSB, a wood particle fraction, disintegration, formaldehyde.

INTRODUCTION

Disposal and treatment of the waste is regulated by the Act No. 79/2015 Coll. on waste and amendments to certain acts valid in Slovakia since January 2016. Wood waste, thus waste of agglomerated wood materials is classified among municipal waste. This material but cannot get on municipal waste landfills if it could be recycled or energy utilised. Accordingly, wood material should not be exported at municipal landfills. One of the reasons is also that the decomposition of this material is environmentally disadvantageous; it damages the environment by harmful degradation products (Rowell et al. 1993, Michanickl 1996, Grigoriou 1996). Recycling of wood waste is difficult due to a content of harmful chemicals contained both in glue used during a manufacture process (Risholm-Sundman and Vestin 2005) and in additives which originally served to protect it from moisture content, wood decaying fungi, to increase fire resistance and so on (Erbreich 2004). Incorporation to new composite materials is one of methods of the recycling. There are many opportunities for composites made from recycled wood-based waste resources. Research in these areas could result in a new product range in combination with other materials that are cost effective, designed to meet demands of end-use, and environmentally friendly (Rowell et al., 1993, Lubke et al. 2014; Ihnat et al. 2015a, b, Wolff and Siempelkamp 2000, Boehme 2003, Saheb and Jog 1999). Agglomerated wood materials are a part of wooden buildings, which are a subject of demolitions at the present. Even this waste it is necessary to address with (Lykidis 2005). Several authors had been devoted to issue of a further processing of large-scale wood-based materials (Sandison 2002, Riddiough and Kearley 2001, Riddiough 2002). Methods of recycling the wood particles from waste wood-based materials were suggested earlier by Sandberg (1965) and later by Michanickl and Boehme (1996).

Products made of wood agglomerates such as chipboards, MDF and structural OSB panels for an energy recovery are mostly to be assigned by the German evaluation (BGBl. I S. 2705 und BGBl. I S 1986) to the second classification group. These materials have been produced using adhesives containing nitrates (urea, melamine), which on the combustion create toxic nitrogen compounds NOX (Salinger 1989, Marutzky 1997). The normative requirement of SR therefore prescribes the specific method of the combustion (Act No. 79/2015 Coll).

Particle boards and MDF are most produced agglomerated materials for furniture industry and OSB panels for a construction use. The boards are produced with a raw surface or surface covered by layers, such as laminates, foils, plastics, veneers according to their use. Ureaformaldehyde (UF), melamine-urea-formaldehyde (MUF), and melamine-urea-formaldehyde resins in combination with a phenolic resin (MUPF) for external environment have been used for boards' production according to the humidity of environment of the use. Formaldehyde used for the resin condensation is gradually released from boards during their lifetime. Formaldehyde is a toxic vapor and its releasing especially from furniture boards is strictly monitored. Just the amount of formaldehyde included in waste agglomerated boards significantly reduces the possibility of their recycling, i.e. their reuse in manufacture of new chipboards and MDF for furniture industry. Therefore, currently used methods of the recycling are based on a maximum reduction of formaldehyde in the recycled chips, respectively fibers.

The currently known method of preparing of particles from agglomerated waste materials according to DE-AS 1201045 was developed by Sandberg (1963). Hydrolysis of adhesives and an associated collapse of wood particles are caused due to water vapour. The method is carried out in a steam chamber under the pressure of 0.1- 0.5 MPa during 0.5 to 4 hours. Produced chips, colored to brown, are seriously damaged by the action of temperature and pressure. Resulting particles may be used for middle layers of particle boards in amount that depends on the level of

residual formaldehyde. But fresh chips must be added to produce particles boards with acceptable properties.

Waste wood agglomerated material is crushed under dry conditions into small fragments which are subsequently treated using steam with 120-180°C at pressure of 0.2 to 1.1 MPa in the patent DE4224629 (1994). Chips created by collapsing of small fragments are heavily damaged by mechanical stress and high temperature. The resulting particles are mixed with native chips and then bonded with binder based on urea.

Impregnation of waste wood with an edge of 10-20 cm by impregnating solution containing urea and sodium hydroxide, or another chemical which causes the alkaline or acidic hydrolysis of old adhesives was described in patent DE 10144793 by Boeme and Michanickl (2003). Impregnation solution must be absorbed by wood material in the amount at least 50% of its weight. In the following second step, the impregnated wood material is heated with steam to temperature of 80 to 120°C in a pressure container. Particles or fibers obtained from the disintegrated wood material are separated by sieving. Obtained chips respectively fiber with a reduced level of residual formaldehyde are suitable for the preparation of new particle boards or MDF by adding of native chips, so that the level of formaldehyde in new boards has been complying with all relevant standards. Riddiough and Kearley (2001) describe the preparation of wood chips using a pressure - vacuum process in the reactor at 180°C. Preparation takes about 1 hour. Particles and fibre obtained contain the residual formaldehyde may be used in the preparation of MDF and chipboards emulsified using non-formaldehyde methylene diphenyl diisocyanate resins for furniture. Disintegration of waste particle boards in dry conditions leads to the formation of high proportion of dust particles (Marutzky 1993).

According to our findings the high proportion of dust particles is created during the dry disintegration and a partial charring of surfaces of larger pieces occurs as well.

MATERIAL AND METHODS

An initial destruction of large dimensions materials was carried out by cutting to smaller pieces approximately 100 x 100 mm. Pallmann drum chipping equipment for manufacturing of chips from natural wood was used for disintegration of these pieces. A solid disintegration mechanism with oval elongated holes ($5.5 \times 54.1 \text{ mm}$), a mechanism with protrusions (opening 4.5 mm, 2.5 mm protrusion) and one with transverse knives (slit- 2.8 x 140 mm) were used. The distribution of prepared particles was determined by the laboratory sieving after drying at 105°C, whereas particles prepared were dried and sieved at pilot plants under the same conditions. The disintegration was performed on two types of waste materials depending on the adhesive used in their production – samples bonded with urea formaldehyde glue (UF) and melamine-urea-formaldehyde glue (MUF).

Disintegration of waste particle boards glued with UF adhesives was carried out on a dry and wet (soaking in water/cooking) material. Experiments on chips boards with a surface foil were provided as well.

a) Disintegration of dry waste particle boards

Pallmann drum chipping semi-production machine with the mechanism with transverse knives (slit- 2.8 x 140 mm) was used to disintegrate samples glued with UF glue with dimensions of 100 x 100 mm and moisture of 8.5%. A large number of burnt dust and small particles smaller

than 5 mm which are not suitable for use was created during the experiment. The disintegration mechanism with oval elongated holes (5.5 x 54.1 mm) was used instead of the mechanism with transverse knives in another experiment. The change caused the burning of produced wood particles which have become not more suitable for further use.

b) Disintegration of wet waste particle boards

Samples of particle boards bonded with UF glues on dimensions of 100 x 100 mm were dipped/ soaked and cooked in water and weighed to determine the water absorbance. The adjustment was provided as 48 hours soaking in cold water and 30 min, 60 min, 120 min and 180 min cooking with stirring (Tab.1). Thus prepared samples were disintegrated and dried at 105°C and particles obtained were sieved to fractions.

c) Disintegration of waste particle boards covered by foil

Waste particle board with a surface foil was overcooked in boiling water (120 min) so as foil could be removed from their tops. The foil was removed along with chips glued to its surface. After removing of foil the sample was processed on Pallmann drum with the mechanism with protrusions (opening 4.5 mm, 2.5 mm protrusion). Particles obtained were characterized by the sieving test. Similarly, other sample has been processed, but that was subsequently disintegrated together with the foil.

Disintegration of waste particle boards and OSB glued with MUF adhesives was provided by mechanical pressing because the samples have remained solid even after 180 min of cooking and a surface burning occurred during their disintegration. Such modified samples of waste particle boards and OSB were subsequently disintegrated using Pallmann drum with the mechanism with protrusions (opening 4.5 mm, 2.5 mm protrusion).

HPLC chromatography system from the company CHROMSERVIS SK Ltd. with two columns - Rezex Pb²⁺ for the determination of formaldehyde and uric acid and Rezex ROA H⁺ for the determination of formaldehyde was used for chemical analysis of samples. Mobile phase consisted of deionised water in the case of the column Rezex Pb²⁺. In case of column Rezex ROA H⁺ the solution of 0.005 N sulphuric acid in deionised water was used. Analyzes on the column Rezex Pb²⁺ were carried out at 80°C and a flow rate of 0.7 ml·min⁻¹. Analyzes on the column Rezex ROA H⁺ were carried out at 30°C and a flow rate of 0.5 ml·min⁻¹. The method of internal standard was used to determine formaldehyde and urea (Fig.1).

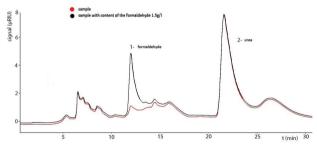


Fig.1: HPLC analysis of the particle board sample by method of internal standard.

RESULTS AND DISCUSSION

Analyzes of large scale agglomerated materials show that different materials behave during their destruction according a type of glue used at their manufacturing. We supposed that the selection of equipment for a primary destruction is closely dependent on equipment for the final disintegration for desired size of wood particles (chips or fibers).

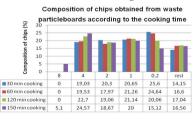
During the disintegration of dry waste particleboards glued with UF adhesives it was found that a surface burning and high dust occur at material with the moisture content of 8.5 %, which was determined by other authors as well (Marutzky 1993). It is therefore necessary that material is swollen with a high content of water. The swelling causes a partial or complete releasing of bonds between wood and adhesives and also the partial hydrolysis of urea-formaldehyde adhesives.

During the disintegration of wet waste particleboards it was found that the relative humidity of samples reaches just only 25 % of water after 48 hours of the soaking in cold water. Boiling water causes significant increase of the water content. The sample begins to break down after 120 min of cooking in boiling water. Total disintegration is achieved after 180 min in boiling water with mixing when the sample reaches the relative moisture content of 66 % (Tab. 1).

Sample	Dry weight (g)	Weight after 48 hours in cold water (g)	Weight after 30 min in boiling water (g)	Weight after 60 min in boiling water (g)	Weight after 120 min in boiling water (g)	Weight after 180 min in boiling water with mixing (g)	Relative moisture content (%)
1	150.14	201.37	-	-	-	-	25.5
2	160.77	-	345.90	-	-	-	53.5
3	153.53	-	-	390.19	-	-	60.6
4	153.96	-	-	-	446.10	-	65.5
5	154.28	-	-	-	-	453.50	66.0

Tab.1: Amount of water in particle boards glued with UF adhesives after the water treatment.

After the subsequent disintegration and fractionation it was found that a very fine material up to 0.5 mm was produced during disintegration using transverse knives of samples processed by water treatment as described above. The sample of particleboard of 100 x 100 mm dipped in cold water during 48 hours began to burn during its disintegration, the attempt was aborted. The soaking in cold water for sample of this size is insufficient for the subsequent processing. Chips from cooked particleboards which were produced using the disintegration mechanism with oval elongated holes (5.5 x 54.1 mm) are characterized in Fig. 2.



Fractions of chips (mm)

Fig.2: Fractions of particleboards after the treatment in boiling water and disintegration using the disintegration mechanism with oval elongated holes (5.5 mm \times 54.1 mm).

Based on the results it can be concluded that:

- soaking in cold water is insufficient for the subsequent disintegration of large particleboard samples
- difference between chips obtained from freely overcooked particleboards and chips from cooked particleboards using the mechanical disintegration is mainly in fractions over 4 mm and 0.2 mm
- the 0.2 mm fraction is about 5-10 % higher for disintegrated samples boiled for 30 min, 60 min and 120 min than for the sample freely overcooked (180 min)
- the total sum of small fractions (0.2 mm and the rest) is in the range of 42- 45 % while for disintegrated samples and only 31.68 % for freely overcooked samples
- the fraction over 4 mm is 10 % higher for overcooked samples than for disintegrated samples
- the fraction over 8 mm does not occurs for disintegrated samples
- the original fractions of particleboards can be obtained by their cooking in boiling water with the permanent stirring
- approximately 30 % of fine particles (usable as surface chips) can be obtained from disintegrated wasted particleboards

Results of sieving tests justify the significant increase in the proportion of fine particles and a significant decrease of particles larger than 4 mm (Tab. 2/sample1) after disintegration of waste particleboards with surface foils. The significant decrease in fractions over 4 mm from 22.70 % to 1.74 % and fractions below 2 mm from 58.24% to 79.53% is detected when the sample boiled for 120 min is compared with the previous case.

Similarly, the sample 2 was boiled for 120 min, which was then disintegrated together with foil using the mechanism with protrusions (Tab. 2/sample2). The resulting chips had the fractional structure more shifted to finer particles than before. Particles above 4 mm are missing and the proportion of particles larger than 2 mm decreased to 9.39 % from initial 18.73 %. The share of particle smaller than 2 mm was 90.61 %. Thus prepared particles are suitable for outer layers. Dimensionally original chips as produced in the manufacture of particleboards are obtained by the overcooking under pressure (Sandberg 1963, 1965, Riddiough and Kearley 2001) or by the chemical attack enhancing the hydrolysis of adhesive (Michanickl and Boeme 2003). Therefore, we also compared chips obtained by our treatment to chips produced by the manufacturer.

Prepared chips have the higher share of fractions of 1 mm and 2 mm compared to fractional composition of particle boards made in real production conditions (Tab. 2/sample3). It appears that a common disintegration of waste particleboards with surface foils would be an appropriate solution of the foil disposal without a separate operation to precede its removal. Resulting particles are suitable for top layers in the manufacture of particleboards.

Waste particleboards with surface foils boiled for 30 min was disintegrated using the mechanism with oval elongated holes (5.5 x 54.1mm). Chips with a higher percentage of fractions 2 mm and 4 mm with values of about 25 % which are achieved in the real production (Tab. 2/ sample 3+4) and suitable for middle layers chips were obtained. The acquired composition of each fraction determined by the sieving test (Tab. 2/sample 5) corresponds to the representation of fractions of particleboards without surface foils (Fig.3). It means that particleboards can be processed with foils in term of the fractional composition of regularly produced particleboards.

Sieve (mm)	Sample 1 (%)	Sample 2 (%)	Sample 3 (%)	Sample 4 (%)	Sample 3+4 (%)	Sample 5 (%)	Sample 6 (%)	Sample 7 (%)	Sample 8 (%)	Sample 9 (%)
8	-	-		1.2	0.85	-	2.36	-	1.68	-
4	1.74	-		34.7	23.51	17.5	31.08	-	28.80	13.11
2	18.73	9.39	1.29	33.7	23.34	19.8	20.09	21.71	23.11	16.94
1	36.26	36.58	16.17	23.1	20.68	21.9	20.29	35.23	22.64	31.05
0,2	34.10	44.52	58.05	5.6	22.61	24.7	21.93	38.46	17.42	33.00
residue	9.17	9.51	24.49	1.7	9.11	16.1	1.25	4.6	6.35	5.90
total	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%

Tab.2: Composition of chips obtained from waste particleboards by cooking and subsequent disintegration.

Notes:

Sample 1 – the particleboard glued with UF resin after the 2 hours cooking treatment without surface foils and subsequent disintegration using the mechanism with protrusions (opening 4.5 mm, 2.5 mm protrusion)
Sample 2 – the particleboard glued with UF resin with surface foils after the 2 hours cooking treatment and subsequent disintegration using the mechanism with protrusions (opening 4.5 mm, 2.5 mm protrusion)
Sample 3 – surface chips for the production of particle boards obtained directly from a production line
Sample 4 – chips for middle layers of particle boards obtained directly from a production line

Sample 3+4 – a recalculated composition of chips for the production of particleboards on production line with 30 % of surfaced chips

Sample 5 – the particleboard glued with UF resin with surface foils after the 30 min cooking treatment and subsequent disintegration using mechanism with oval elongated holes (5.5 x 54.1 mm).

Sample 6– the laminated particleboard glued with MUF resin after the 180 min cooking treatment, crushing in press and subsequent disintegration using mechanism with oval elongated holes.

Sample 7 – the laminated particleboard glued with MUF resin after the 180 min cooking treatment, crushing in press and subsequent disintegration using a mechanism with protrusions (opening 4.5 mm, 2.5mm protrusion)

Sample 8 - OSB glued with MUF resin after the 180 min cooking treatment, crushing in press and subsequent disintegration using a mechanism with oval elongated holes.

Sample 9 – OSB glued with MUF resin after the 180 min cooking treatment, crushing in press and subsequent disintegration using a mechanism with protrusions (opening 4.5 mm, 2.5 mm protrusion)

Disintegration of waste particleboards glued with MUF adhesives was conducted using the mechanism with oval elongated holes. When comparing the different fractions, most chips are retained by the 4 mm mesh (31.08%). There are 26.18 % of particles smaller than 1 mm and 55.53% of fractions smaller than 2 mm in the composition (Tab. 2/sample 6). Fractions above 2 mm were formed by overcooking of waste particleboards bonded with UF resins, which represents 48.34%. That is 5.19% less than in case of boards to environment with an increased humidity. It is obvious that the structure of chips produced by manufacturer is different; furniture boards have a greater percentage of fine particles in surface (31.69% vs. 26.18%). Particles obtained are suitable for manufacturing of particleboards and chips above 2 mm for a fibre manufacture. Chips, obtained using the mechanism with protrusions, have the finer composition as above. The sieving confirmed the absence of chips over 4 mm (Fig. 2/sample 7). The mechanism with protrusions is not suitable equipment for the material preparation for the next fiber production due to the high amount of particles less than 2 mm (78.29 %). The particles obtained after the sieving above 2 mm could be suitable for a production of surface layers of particles obtained after the

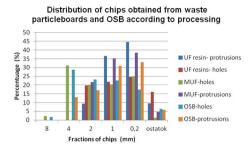


Fig.3: Composition of chips from particleboards and OSB after the cooking treatment and the subsequent disintegration using the mechanism with protrusions.

Chips obtained by disintegration of OSB glued with MUF adhesives using the mechanism with oval elongated holes (Fig. 2/sample 8) have a similar composition to laminated particleboards glued with MUF resins (Fig. 2/sample 6). Chips above 2 mm represent 53.59% in case of OSB, in case of the comparative particleboard it is about 54.53%. The chips made by disintegration of OSB are suitable for the manufacture of particleboards; just particles above 2 mm are suitable for the MDF pulp production. Chips prepared with the mechanism with protrusions (Tab. 2/sample 9) contain 69.95% of particles below 2 mm, which is the greater percentage of fine particles than in the case of the oval elongated holes (46.41%). The chips could be used for surface layers of new particleboards. The composition of chips obtained from waste particleboards and OSB after the cooking treatment and subsequent disintegration is shown in the graph (Fig. 3).

Determination of formaldehyde and urea using HPLC in waste water after the cooking treatment of waste particleboards before their disintegration

The original amount of formaldehyde and urea contained in particleboards gradually decreases due to a hydrolysis of UF adhesives. The evaluated amount of 100 g of the absolutely dry mass of particleboards contained 3.225 g of formaldehyde and 5.860 g of urea. The amount of formaldehyde emissions depends on cooking time and size of the particleboard sample (Fig. 4). Process of the formaldehyde emission is the slower for large samples due to the hydrolysis of resins during the cooking treatment. The 30 min cooking treatment caused the hydrolysis of 20 % UF resins.

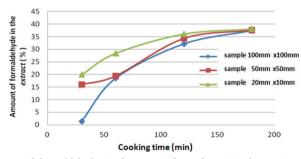


Fig.4: The percentage of formaldehyde in the extract from the original quantity of formaldehyde in particleboards.

It is expected that the amount of formaldehyde in prepared chips obtained from waste particleboards after the cooking treatment will be reduced at least by values shown in Fig.4. But the amount of formaldehyde in the extract is just an indicative parameter. The amount of formaldehyde in newly prepared particleboards will be important.

CONCLUSIONS

To prepare chips and pulp for the production of new particleboards and MDFs from waste particleboards and OSBs the following findings are important:

- Dust and the burning of particles with 10 cm edges appear during disintegration of the dry waste agglomerated material on the drum machine Pallmann.
- The 48 hours dipping in cold water of samples with dimensions 10 x 10 cm is insufficient because the material achieves just 25% of its relative moisture content and the burning of its surface still appears during disintegration.
- The 180 min cooking of wasted particleboards disintegrates the boards to origin particles.
- Difference between chips obtained from freely overcooked particleboards and chips from cooked particleboards using the mechanical disintegration is reflected mainly in fractions over 4 mm and 0.2 mm.
- Total sum of small fractions (0.2 mm and smaller) is in the range of 44-45% for disintegrated samples and only 31.68 % for freely overcooked samples. The fraction over 4 mm is 29.67% for overcooked samples and in range 19-22% for disintegrated samples. The fraction over 8 mm does not occur for disintegrated samples.
- Waste particleboards and OSB with dimensions 100 mm x100 mm bonded with MUF adhesive for the exterior use should be treated by cooking followed by the mechanical crushing. Such modified particleboards and OSB can be disintegrated using Pallmann.
- Waste particleboards and OSB glued with MUF treated in the way described in previous clause using the oval elongated openings have chips composition suitable for the production of particleboards.
- Waste particleboards with foils treated in this way and disintegrated using the mechanism with protrusions have chips composition suitable for top layers of particleboards.
- It is assumed that the amount of formaldehyde in the chips prepared from waste particleboards after the cooking treatment is reduced at least according to the values shown in Fig. 4.

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