Quality characteristics of hydrothermally recycled particleboards using various wood recovery parameters

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The aim of the present paper was to study the effect of some particle recovery parameters on the quality of hydrothermally recycled particleboards. The research was carried out in two phases: rough evaluation of various recovery parameters and determination of the recycled particleboard properties. It was concluded that the optimum group of hydrothermal recovery parameters (among those tested in this investigation) were 45% water retention/150°C temperature/10 min duration. This conclusion relies on the below-mentioned facts: the conditions of recovery parameters 45%/150°C/10 min resulted in the lowest agglomeration ratio in the recovered material in relation to the other recovery parameters examined in the first and second phases. Concerning the properties of the recycled particleboards, it was observed that the boards that were produced utilising the above-mentioned recovery parameters showed the best internal bond, surface soundness, modulus of elasticity in bending, hygroscopic properties and free formaldehyde content values.

Keywords: Hydrothermal treatment, Particleboard, Recovery, Recycling, Wood, Recycled particleboard properties

Introduction

It is generally recognised, not only within the scientific community but also in general, that the natural environment is currently undergoing very strong pressure due to various factors, such as air pollution, contamination of underground water reserves, significant reduction in forest area and degradation of terrestrial and aquatic ecosystems, the latter of which are mostly expressed by municipal and industrial waste. Municipal waste includes various types of materials including paper, cardboard, glass, rubber, leather, textiles, plastics, metals and wood. The waste wood represents a considerable part of urban waste and consists of wood furniture, cabinets, pallets, packaging, sawn timber and glued wood products residues, waste from manufacturing plants as well as other products (Jungmeier et al. 2005).

The efficient reuse and recycling of wood waste presents an opportunity to extend timber resources, reduce consumption of new resources, reduce landfills, reduce cost through avoided purchase/disposal fees, preserve carbon storage, reduce energy and create 'green' jobs (Bratkovitch *et al.* 2009).

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In recent years, wood-based panel industries have been trying to intensify their efforts regarding sustainable forest management by increasing the amounts of recovered wood used in the manufacture of their products (EPF 2003, 2005). Particleboard industries for many years have been using waste particleboards either from their own production or from other sources (old wood constructions). The discarded material is crushed and driven in the production line of new boards. The result of this practice presents disadvantages concerning the geometry of the recovered wood particles and thus quality deterioration of the recycled boards (Czarnecki et al. 2003). As a result of this unfavourable influence, the industries tend to use a limited amount of recovered particles, namely, in the core layer of the boards (Boehme 2003; Kearley et al. 2005).

The above problem can be faced by recovering wood particles/fibres through hydrothermal treatments. Under the influence of water and heat, it is easy to hydrolyse the urea–formaldehyde adhesives which are used for the production of particleboards and medium density fibreboard and to detach the woody materials, which will then be available for the production of new (recycled) particleboards/fibreboards (Pfleiderer Unternehmens-verwalt 1994; Boehme and Michanickl 1998; Roffael *et al.* 2002; Kearley and Goroyias 2004).

Owing to the above recovery procedure, these methods present the advantage that after the recovery process, wood particles carry residual adhesive which is re-activated and helps both reduce consumption of new adhesives (Nakos and Roffael 1998) and also reduce formaldehyde release from the recycled boards (Roffael

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and Franke 1995; Michanickl 1996a; Lykidis and Grigoriou 2008). Another advantage of these methods is the efficient removal of other materials used in the manufacture of various wood constructions, such as metal parts (handles, hinges, etc.) and other non-woody materials (plastics, glass, etc.) that should not be introduced into production of new boards because of problems related to the deterioration of processing and production machinery of recycled boards.

Boehme and Michanickl (1998) patented a recovery methodology referred to as the multi-stage, chemothermomechanical method. In this methodology, waste particleboards are crushed, impregnated with an aqueous solution consisting of water, urea, ammonia and other chemicals and then heated at 80-120°C (Boehme and Michanickl 1998; Boehme 2003; Michanickl and Boehme 2003). Results of the above methodology were presented by Michanickl (1996a,b) who stated that recovered particles can be used in manufacturing particleboards mixed with fresh particles without causing noticeable differences in board quality. According to Riddiough and Kearley (2001) although the above methodology appears to be effective in terms of recycled board quality, it presents the drawback of the load of chemical compounds, such urea, ammonia and soda, which, as mentioned by the authors, are necessary to recover wood particles.

Alpar *et al.* (2007) conducted research in which crushed particleboards were exposed to hydrothermal treatments at temperatures in the range of $100-180^{\circ}$ C and duration of 10-20 min. To identify the changes that occurred in cells of recovered particles, they compared them microscopically with cells of particles obtained from crushed particleboards (without hydrothermal treatment). An important finding of this comparison was that the recovered particles showed reduced distortion compared with particles from crushed particleboards, a finding which was attributed to the hydrothermal treatment by the researchers.

Lykidis and Grigoriou (2008) investigated the effects of hydrothermal treatment parameters utilised for the recovery of wood particles from particleboards on the properties of the recycled particleboards. Specifically, laboratory-produced boards were hydrothermally treated in four different conditions of pressure-temperature-time for recovery of particles. The utilised conditions were: 2 bar/119°C/480 min, 4 bar/140°C/120 min, 6 bar/156°C/ 45 min and 8 bar/167°C/20 min. The hydrothermal treatments were carried out without pre-impregnation of the particleboards with any aqueous solution. The recovered particles were used for the production of recycled boards. The results showed that in the case of some hydrothermal conditions the recovered material included limited numbers of agglomerated particles. It was also found that the recycling of particleboards through hydrothermal treatments adversely affects the quality of recycled boards in regard to both the mechanical and hygroscopic properties [except the modulus of elasticity (MOE) in bending].

Taking into consideration the results of the above investigation, the aim of the present paper was to study the effect of new particle recovery parameters on the quality of hydrothermally recycled particleboards. In contrast to the methods described above, the methods used in this paper involved combinations of water impregnation – at different retention ratios – of the



1 Parameters investigated in terms of their effect on quality of recovered material

waste particleboards before the hydrothermal treatment as well as the application of a vacuum. The percentage of agglomerated particles that occurred in the recovered material after the application of the various recovery methodologies was also studied. The research was carried out in two phases. The first aimed at roughly evaluating various recovery parameters using as a criterion the quality of the recovered material, and the second aimed at determining the best of the combinations in terms of the quality characteristics of the recycled particleboards.

Experimental method

In conducting the first and second research phases, commercially produced particleboards, covered with white melamine impregnated paper, of the same quality were used. These particleboards were produced under the same production parameters, which was necessary in order to achieve comparability between the different recovery methods.

The hydrothermal recovery treatments were carried out in a laboratory scale device having the capability to produce saturated steam at a temperature range of $100-175^{\circ}$ C, impregnate the waste material with liquid impregnation material and pressurise with air in the range of 1–8 bar (absolute) and apply vacuum in the range of 1–0.03 bar (absolute).

Figure 1 presents the recovery parameters which were studied in terms of the quality of the recovered material. More specifically, the quality criterion used in the first phase was the percentage of agglomerated particles present in the recovered material. The increased percentage of agglomerates in the recovered particles is an indication of incomplete resin decomposition during the recovery process, and possibly results in low recycled board performance in terms of mechanical and hygroscopic properties.

As can be seen in Fig. 1, the temperatures utilised in the first phase were 110, 130 and 150°C. Utilisation of higher temperatures was avoided due to the significant thermal decomposition that occurs to the wood components and which is projected as a significant decrease in the mechanical properties of the recycled boards (Lykidis and Grigoriou 2008). Concerning the impregnation of the waste boards, the impregnating solution chosen was water, and the retention ratios tested were 0, 30, 45 and 60% (based on the weight gain of the boards).



2 Methodology steps carried out for production of recycled particleboards

Another parameter studied was the utilisation of a vacuum (20 mbar absolute) after the hydrothermal treatment. Also studied was a second treatment cycle of the performed treatments. For every combination of the above tested parameters, four repetitions of recovery treatments were performed. In order to find the optimal combination, the duration of the hydrothermal treatments varied between 8 and 180 min for every combination of the above tested parameters. The optimum treatment duration, for every combination of recovery parameters, was identified as the minimum duration needed in order to achieve proportions of agglomerated particles in the recovered material below 10%. After the assessment of the result of the first research phase, seven groups of particle recovery parameters with optimal duration for each one were chosen.

The second research phase (Fig. 2) aimed at the detailed comparison of the above seven groups of parameters in order to find the optimal one. To do this, the seven groups of recovery methods were used to recover particles from the same type of commercial boards used in the first phase. After the treatments, the surface coatings were separated using a proper mesh and were not used in the production of recycled particleboards. This decision was made in order to facilitate the accurate comparison of the seven different recovery parameters by reducing the factors influencing the quality of the recycled boards. The seven different recovered materials were used for the production of laboratory particleboards of the following characteristics: thickness of 12 mm, density of 0.68 g cm^{-3} , threelayered (surface to core layer weight ratio of 40:60). The

Table 1 Moisture content and agglomeration ratios of recovered materia	Table 1	Moisture	content and	agglomeration	ratios of	of	recovered	materia
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Recovery parameter		Moisture content of the recovered material/%	Agglomeration ratio (in the recovered material) C _p /%	Agglomeration ratio (in the core layer of the recycled boards) $C_r/\%$			
1	30%/150°C/20 min	37.5	9.14	15.23			
2	45%/110°C/75 min	69.4	4.97	8.29			
3	45%/130°C/20 min	68.8	1.45	2.41			
4	45%/150°C/10 min	70.3	0.57	0.95			
5	60%/110°C/30 min	85.6	3.34	5.56			
6	60%/130°C/10 min	83.8	1.84	3.07			
7	60%/150°C/8 min	83·6	1.46	2.44			

recovered particles were sieved with a 1.5 mm mesh and the material with dimensions less than 1.5 mm was used in the surface layer, while the material with dimensions over 1.5 mm was used in the core layer of the produced boards. The resin used for the production of the laboratory boards was a commercial urea–formaldehyde resin E2 class (according to EN 13986:2004), and it was used at a dry weight ratio of 8% for the core and 12% for the surface layers. The hardener used was ammonium chloride at a dry weight ratio of 2% (per dry resin weight). The hot pressing temperature was 185°C and the total pressing duration was 240 s (30 s to reach maximum pressure, 150 s at maximum pressure and 60 s for degassing).

For all types of particleboards produced, determination of density, internal bond (IB), surface soundness (SS), modulus of rupture (MOR) in bending, MOE in bending, thickness swelling (TS), absorption of water (WA) within 24 h and formaldehyde content (perforator method) were determined according to the current European Norms (EN 120:1992; EN 311:1992; EN 310:1993; EN 317:1993; EN 319:1993; EN 323:1993). The results for each property were expressed in mean and standard deviation values. Statistical analysis of variance with a confidence level of 95% was performed to determine whether significant differences among the mean values of the tested parameters occurred.

The recovered particles that were produced after the seven different groups of treatments included a portion of agglomerated particles, which obviously resulted from the incomplete resin decomposition throughout the hydrothermal treatments. The percentage of the particle incorporations in the recovered material was assessed using the formula below

$$C_{\rm p} = \frac{m_{\rm c,0}}{m_{\rm p,0}}$$
(1)

where $C_{\rm p}$ is the percentage of dry particle agglomerations present in the dry mass of hydrothermally treated particleboards (%), $m_{\rm c,0}$ is the dry mass of particle agglomerations present in the recovered material (g) and $m_{\rm p,0}$ is the dry mass of hydrothermally treated particleboards (g).

The recovered, agglomerated particles were used only in the core layer of the laboratory boards, as they did not pass through the 1.5 mm mesh. Additionally, to determine the effect of the agglomerations on the properties of the recycled boards, the weight of the agglomerations was also calculated as a percentage of the core layer material of the recycled boards.

The calculation was made using the formula below

$$C_{\rm r} = \frac{100 \times m_{\rm c,0}}{60 \times m_{\rm r,0}} \tag{2}$$

where $C_{\rm r}$ is the percentage of dry particle agglomerations present in the dry mass of the core layer of the recycled boards (%), $m_{\rm c,0}$ is the dry mass of particle agglomerations present in the recovered material (without the coatings) (g), $m_{\rm r,0}$ is the dry mass of the recovered material (without the coatings) (g) and 60 is the core layer ratio of the laboratory particleboards.

Results and discussion

According to the research, the following seven combinations of particle recovery parameters presented in Table 1 (impregnation rate/temperature-pressure/time)

recycled boards	Recovery parameter		Thickness/ mm	Density/ g cm ⁻³	IB/ N mm ⁻²	SS/ N mm ⁻²	MOR/ N mm ⁻²	MOE/ N mm ⁻²	TS/%	WA/%
1	30%/150°C/20 min	Average (s)	11·84 0·0621	0·69 0·0352	0·38 0·1078	0·87 0·1626	13·53 1·6152	2686·87 251·8562	21·80 0·0260	80·05 0·0561
		Max.	12·00	0.78	0.20	1.15	17.80	3065.13	28·55	91·43
		Min.	11.73	0.61	0.16	0.60	10.66	2110.86	17.22	69.78
2	45%/110°C/75	Average	11.89	0.68	0.43	0.90	12.95	2248.11	20.83	84·39
	min	(s)	0.0398	0.0326	0.0578	0.1012	1.1778	192.4666	0.0106	0.0679
		Max.	11.98	0.76	0.57	1.05	14.82	2627.51	22.20	99.50
	150/110000/00	Min.	11.79	0.60	0.35	0.69	10.35	1990.90	18.66	74·01
3	45%/130°C/20	Average	11.79	0.70	0.53	0.98	13.46	2339.86	20.30	81.44
	min	(S)	0.1320	0.0283	0.0443	0.1036	1.5316	144.2660	0.0124	0.0404
		Max.	11.96	0.75	0.61	1.17	16.49	2555.65	22.47	87.45
	450/ 145000 140	Min.	11.55	0.63	0.45	0.75	10.86	1997.60	1/./1	73.30
4	45%/150°C/10	Average	11.76	0.69	0.54	0.99	14.16	2534.59	19.64	/9.//
	min	(S)	0.1317	0.0286	0.0317	0.1237	1.4300	281.1056	0.0152	0.0410
		Max.	11.92	0.74	0.58	1.51	10.37	3139.64	22.59	88.10
F	600/ /110°C/20	IVIIII.	11.00	0.64	0.45	0.73	11.60	2129.35	16.20	7 1.68
5	60%/110 C/30	Average	0 1400	0.0294	0.0677	0.70	9.21	1/02.0/	23.99	00.32
	111111	(S) Mox	11.07	0.76	0.0077	0.07 10	0.9420	147.1204	0.0100	0402
		Min	11.97	0.76	0.69	0.69	10.66	2122.79	20.02	94·30 70.72
6	60% /120°C/10	Avorago	11.95	0.69	0.46	0.05	10.00	2110.00	10.96	79.73 92.12
0	00 %/ 130 C/ 10	(s)	0.0424	0.0249	0.0508	0.0028	1.3255	161,6308	0.0134	0.0454
	111111	(S) Max	11.05	0.75	0.52	1.1/	15.64	2576.00	22.34	01.83
		Min	11.78	073	0.34	0.77	10.50	1969.97	17.85	72.49
7	60%/150°C/8	Average	11.85	0.69	0.54	1.02	13.92	2448-21	19.37	79.90
	min	(s)	0.0381	0.0349	0.0692	0.1198	1.3424	164.0429	0.0180	0.0432
		Max	11.96	0.77	0.64	1.28	15.69	2703.98	22.51	87.22
		Min	11.78	0.61	0.38	0.85	10.99	2179.94	16.92	70.23
		n	60	60	18	36	15	15	18	18

Table 2 Properties of recycled boards

16.00 14.40 14.00 Perforator value (mg/100g) 12 64 12.00 10.75 9.92 10.00 E2 7.56 8.00 6 15 E1 6.00 3.68 4.00 2.00 0.00 30%/ 150°C/ 45%/ 110°C/ 45%/ 130°C/ 45%/ 150°C/ 60%/ 110°C/ 60%/ 130°C/ 60%/ 150°C/ 20min 75min 20min 10min 30min 10min 8min 2 3 4 5 6 7 1

Formaldehyde content (Perforator)

3 Formaldehyde content (perforator values) of laboratory particleboards

showed agglomeration rates (dry mass of agglomerations per dry mass of particleboards) below 10%: no. 1 $(30\%/150^{\circ}C/20 \text{ min})$, no. 2 $(45\%/110^{\circ}C/75 \text{ min})$, no. 3 $(45\%/130^{\circ}C/20 \text{ min})$, no. 4 $(45\%/150^{\circ}C/10 \text{ min})$, no. 5 $(60\%/110^{\circ}C/30 \text{ min})$, no. 6 $(60\%/130^{\circ}C/10 \text{ min})$ and no. 7 $(60\%/150^{\circ}C/8 \text{ min})$. These seven groups of parameters were used in the second research phase in order to evaluate the optimum among them.

Table 1 lists the moisture content and agglomerations of the recovered particles for each of the groups of conditions that were selected in the first phase. The above table implies that the water retention rate of the particleboard specimens before the hydrothermal treatment considerably influenced the final moisture content of the recovered material. In particular, higher retention rates lead to higher moisture content of the recovered material. It can also be derived from Table 1 that with the exception of one case, higher recovery temperatures result in lower amounts of particle agglomerations. The lowest agglomeration rates were achieved utilising the following parameter groups: 45%/130°C/20 min, 45%/150°C/10 min and 60%/150°C/8 min.

The properties of recycled particleboards made of recovered materials after the application of the seven groups of recovery conditions are presented in Table 2.

As can be seen in Table 2, the densities of laboratory particleboards varied within the range of 0.68- 0.70 g cm^{-3} , and according to the statistical analysis no significant differences were found among them. This fact provides an adequate basis for objective comparison between the recycled particleboards. Taking into consideration the data of Table 1, it seems that - with the exception of the recovery parameters no. 5 - decreased amounts of particle agglomerations in the core laver positively affected the IB values of the particleboards. In addition to the above, it can also be concluded that the soaking of particleboards at water retentions over 30% significantly increases IB values of the recycled laboratory particleboards. This finding has been also reported by Michanickl (1996b) and Michanickl and Boehme (1996). According to Table 2, among the recycled boards those produced with recovery parameters no. 3, no. 4, no. 5 and no. 7 showed statistically significant improvement in IB values compared with those produced with the rest of the recovery parameters. Moreover, parameters no. 3, no. 4 and no. 7 resulted in the highest SS of the recycled boards with statistically insignificant differences between them, while parameters no. 1, no. 2, no. 5 and no. 6 resulted in significantly lower values of the same property.

Concerning the MOR (Table 2), the highest values of the recycled boards were achieved by the recovery parameters no. 1, no. 3, no. 4 and no. 7; no statistically significant differences were found among them. The rest of the recovery parameters resulted in a significantly lower MOR of the recycled boards. Generally, the MOE values of the recycled boards corresponded to those of MOR; an unexpected finding was that the boards recycled utilising recovery parameters no. 1 showed statistically significant higher values of MOE.

The recovery parameters that resulted in the lowest TS of the recycled boards after 24 h immersion in water were no. 3, no. 4, no. 6 and no. 7, with statistically insignificant differences among them. Significantly higher values of this property were determined for the recycled boards of the recovery parameters no. 1 and no. 5. Additionally, the recovery parameters that resulted in the lowest WA of the recycled boards after 24 h immersion in water were no. 1, no. 3, no. 4 and no. 7, with statistically insignificant differences among them. The recycled boards of the recovery parameters no. 2 and no. 5 showed significantly higher values of this property. The trends observed in the water absorption of the recycled particleboards were in line with the TS.

Figure 3 shows the values of formaldehyde content of the recycled particleboards compared with the EN 312:2003 marginal values for the E1 class. The above figure suggests that the boards that showed the lowest formaldehyde content values were produced applying the recovery parameters no. 1, no. 4 and no. 7.

It is also noteworthy that these formaldehyde content values belong to E1 class, although the recycled boards were produced using E2 urea–formaldehyde adhesive. Similar findings have also been reported in other relevant studies (Roffael and Franke 1995; Michanickl 1996a,b; Dix *et al.* 2001a,b; Lykidis and Grigoriou 2008). The explanation for this lies in the presence of urea and other derivatives of hardened urea–formaldehyde degradation such as dimethyl-urea (Roffael and Kraft 2005). These substances found in the recovered material are activated during the production of recycled particleboards and act as scavengers (catchers) of the excess formaldehyde. It is likely that the higher temperature (150°C) of hydrothermal treatments of the recovery parameters no. 1, no. 4 and no. 7 in comparison with the lower temperatures (110 and 130°C) of the other parameters leads to increased adhesive degradation of the recovered boards and therefore promotes the activation of urea as formaldehyde scavenger.

Conclusions

From the above discussion of the results, it can be concluded that, basically in terms of the quality of recycled boards, the optimum (among those tested in this investigation) hydrothermal recovery parameters were 45% water retention/150°C temperature/10 min duration. This conclusion relies on the following facts: the conditions of recovery parameters no. 4 (45%/150°C/ 10 min) resulted in the lowest agglomeration ratio in the recovered material in relation to the other recovery parameters examined in the first and second phases. Concerning the properties of the recycled particleboards, it was observed that the boards that were produced utilising recovery parameters no. 4 along with those of recovery parameters no. 3, no. 5 and no. 7 showed the highest IB values. With regard to the SS, recovery parameters no. 4 along with no. 3 and no. 7 resulted in the highest values of recycled boards. Regarding the MOR and the MOE in bending, the boards that were produced utilising the recovery parameters no. 4 together with no. 1, and no. 7 showed the highest values. Concerning the hygroscopic properties of the recycled boards, parameters no. 4 as well as no. 3, no. 6 and no. 7 showed the best quality. In addition, laboratory boards corresponding to parameters no. 4 along with parameters no. 1 showed the lowest free formaldehyde content.

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