Utilisation of Coloured Paper Refuse in Eco-products

Alexandra TAKÁTS[a*] − Péter TAKÁTS[b]

[a] Institute of Informatics and Economics, Faculty of Wood Sciences,
University of West Hungary, Sopron, Hungary
[b] Institute of Wood and Paper Technology, Faculty of Wood Sciences,
University of West Hungary, Sopron, Hungary

Abstract – In the course of our research we have found a solution for recycling coloured advertising paper in a cellulose fibre based composite. This paper quality is produced in huge quantities and can be recycled with difficulties even in paper industry. Our aim was to produce a kind of organic composite that does not contain any adhesives fixing the matrix part of the composite. An eco-product generated this way has similar physical and mechanical attributes to medium density fibreboard (MDF). Fibre made of coloured newspaper can easily be adapted to board production while, at the same time, this supports also the development of an environment oriented product policy. Board-based composites produced by utilizing recycled refuse paper can be used for designing and creating environmentally friendly products following the eco-design trend.

refuse paper / fibre composite / free from adhesive / eco-product / environment-oriented product policy / eco-design

Kivonat – Papírhulladék hasznosítása ökotermék előállítására. A kutatómunka során a nagy mennyiségben keletkező és papíripari célra is nehezen újrahasznosítható színes reklámújság hulladék cellulózrost alapú kompozitban történő hasznosítására sikerült megoldást találnunk. A cél egy olyan biokompozit termék előállítása volt, melynek gyártása során a kompozit mátrix részét biztosító külön kötőanyag nem került felhasználásra. Az így elkészített ökotermék a közepes sűrűségű farostlemezhez (MDF) hasonló fizikai-mechanikai tulajdonságokkal rendelkezik. A színes újságpapírból készült rost a lapgyártás folyamatába könnyen beilleszthetővé válhat, miközben jelentős környezeti terheléstől képes közvetlenül megszabadítani a természetet, lehetővé téve ezzel egy környezetorientált termékpolitika létrejöttét. Az újrahasznosított hulladékpapír felhasználásával készült lapalapú kompozitok így alkalmassá tehetők környezetbarát termékek tervezésére, kialakítására egy ún. ökodesign irányvonal bevezetése révén.

papírhulladék / rostkompozit / kötőanyagmentes / ökotermék / környezetorientált termékpolitika / ökodesign

1 INTRODUCTION

The aim of environment oriented product policy is to find the versions providing optimal protection of the environment by taking into account the whole life cycle of products. A comprehensive concept is needed that includes the planning, the manufacturing and

[*] Corresponding author: sztakats@fmk.nyme.hu, H-9400 SOPRON, Bajcsy-Zs. u. 4.

eventually the disposal or recycling of a product. Eco-design in this sense is a preventive planning process with environmental protection in the centre.

The problem of refuse materials is one of the widest and most diversified problems of environmental protection. While the appearance of refuse materials is inevitable, the differentiation between refuse material disposal and refuse material recycling is about to disappear parallel with the advancement of ideas about the refuse problem (Takáts 1998).

Refuse paper provides not only recyclable material to produce paper again but represents also a huge potential raw material base to be used in the composite technologies *(Figure 1).*

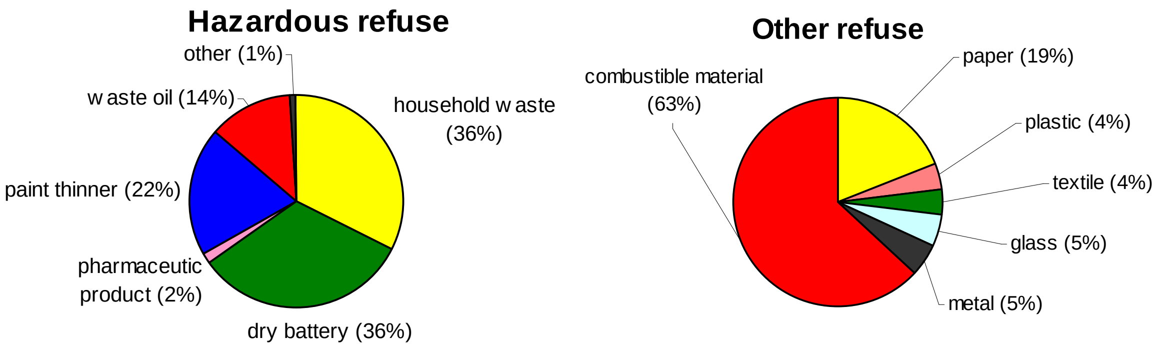


Figure 1. Average composition of communal solid refuse in Hungary (2000–2005) (KVVM 2006)

Paper industry was among the first to realise the importance of environmental protection and the reasonable management of natural resources in the last 30 years. As a result of this, cellulose companies have become net energy emitters and paper manufacturers have reduced their fibre needs to 50%, their specific energy consumption to one-third and their water consumption to one-tenth. For example, the recycling of paper reached a record of 32 million tons in Europe in 1996, one-third of which (more than 10 million tons) was collected in Germany. The ratio of recycling was 49.8%. According to the data of CEPI (Confederation of European Paper Industries) the ratio of recycled paper used in paper production has increased by 10% in Europe in the last 10 years. With this recycling rate, paper industry surpasses all other industrial branches.

Secondary fibre uses are even more favourable in the Hungarian paper industry than the EU average due to its product composition (high proportion of wrapping paper, card board, and hygienic paper). A significant part of refuse paper need is covered from import. However, collection ratios have considerably improved in the last ten years (Figure 2.). It has to be mentioned that the increase in the ratio of recycling has its limits. During manyfold recycling, fibres get shorter and the quality of paper product decreases. More emphasis has to be put on preserving sound fibres by moderate methods during fibrification (Winkler 1999).

Recycling of paper does not reach the level required by environmental protection or recycling interests in the world, although countries that consume great quantities of paper recycle a significant amount of it. The general difficulty in recycling refuse paper: paper manufacturing companies have high requirements for refuse paper to be returned into the production process. That is why we have to search for other opportunities in recycling refuse paper. One alternative could be the application in wood industry, especially in the furniture industry, by producing new eco-composite products.

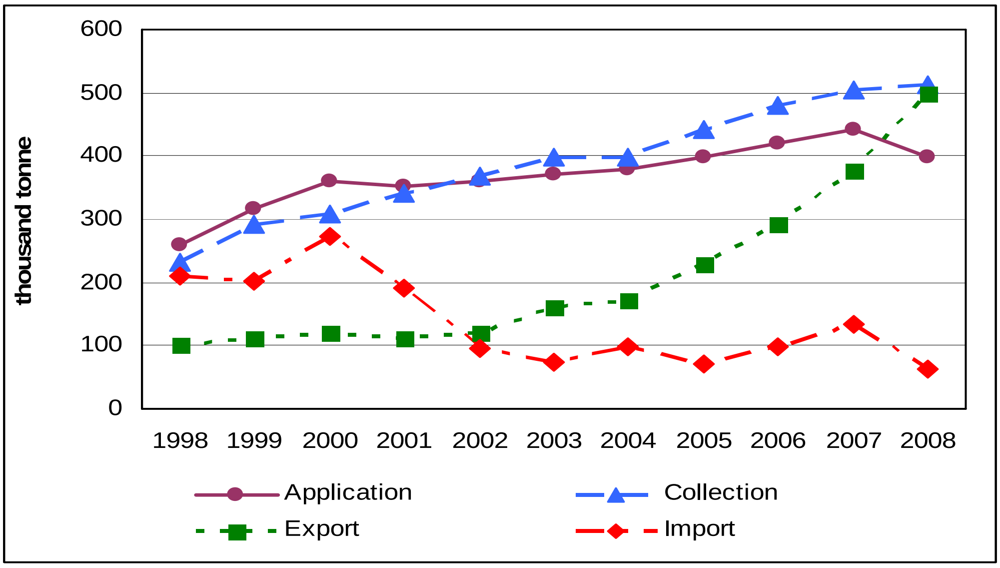


Figure 2. Refuse paper turnover in Hungary (1998–2008)
(Kopint-Datorg 2010)

The highest quality refuse paper can be produced by selective paper collection. Selective paper collection is considered to be a new phenomenon for the population in Hungary. People still have to be made aware of this process and they have to realise the fact that this activity will have visible results in the reduction of environmental burdens in the long-run. The deposition of refuse containing printer's ink extracted from refuse paper still has to be solved without harming the environment. The recycling provides a solution for the application problem which spares this activity of great costs.

The use of refuse paper as secondary raw material opposes the interests of waste incineration. The seemingly economical process of burning needs an ever-increasing heat content in the dry refuse. Secondary raw material use extracts one of the components with high heat content from mixed communal refuse. However, the recycling of paper is twice as economical as burning.

Types of refuse paper

In the European Union CEN (Centre Européene de Normalisation, Bruxelles) created the European bill of standardised qualities of refuse paper in 1994 amalgamating the experiences of individual nations. Hungary introduced the bill in the following year despite the fact that it was not the member of the EU at that time.

BIR (Bureau of International Recycling, Brussels) and CEPI (Confederation of European Paper Industries, Brussels) prepared the modernisation of the above-mentioned standard by 1999. This enables a five-level classification of refuse paper and groups refuse paper according to its origin and type into the following categories: mixed-low quality (A), moderate (B), good quality (C) and containing only unbleached cellulose (kraft) (D). In addition to the fourth category of unbleached fibre refuse paper, this standardisation creates a fifth group: special refuse paper requiring unique technologies (E) *(Table 1).*

Table 1. Refuse paper categories as suggested by CEPI and BIR

Refuse paper of mixed quality (A)	**Refuse paper of moderate quality (B)**	**Good quality refuse paper (C)**	**Unbleached fibre refuse paper (D)**	**Special refuse paper (E)**
Mixed 1, uncategorised	Newspapers with max. 5% of coloured parts inside	Lightly coloured, mixed edge refuse	Corrugated box – factory refuse	Mixed paper and card board
Mixed 2, categorised	Unsold newspapers, no coloured parts inside	Bindery refuse***	Corrugated box – factory refuse (celluloses and semi-celluloses);	Mixed wrapping paper types;
Card board	Unsold newspapers, no coloured parts inside or flexo-pressure	Torn, white refuse	Corrugated box – factory refuse (kraft- and test liner)	Liquid containing boxes
Corrugated cardboard from stores	Edge refuse from printing house, little printer's ink	White writing***	Used corrugated boxes I (only kraft-liner and semi-celluloses fluting)	Kraft packing paper
Used corrugated cardboard	Edge refuse from printing house, little printer's ink *	White business publication	Used corrugated boxes, minimum one kraft-liner layer;	Wet-solid label paper
Unsold periodicals	Edge refuse from printing house, printed	Computer publications ***	Used sacks;	Unprinted, white, wet-solid paper
Unsold periodicals*	Edge refuse from printing house, printed *	Printed white celluloses card board	Used sacks with PE cover	Printed, white wet-solid paper
Phonebook	Office papers, classified	Slightly printed white celluloses card board	Unused sacks	
Newspaper (min.50%) and periodical I.	Coloured writing	White, painted publication***	Unused sacks with PE cover	
Newspaper (min.60%) and periodical II.	White book***	White, printed cardboard	Used kraft papers	
Periodical (min. 60%) and newspaper*;	Coloured periodical***	White, slightly printed card board	Unused kraft papers	
Public brochures**	Carbon free copier	White, unprinted card board	Unused shopping bags	
	White PE covered cardboard	Unprinted newspaper		
	PE covered card board***	Bleached, wood containing, painted or unpainted, unprinted paper		
	Fat. computer publications	Bleached, wood containing, painted paper, unprinted		
		White painted paper, unprinted**, ***;		
		White edge refuse; unprinted		
		White edge refuse; unprinted ***;		
		Unprinted, white celluloses card board		

Note: Fat.: contains wood, * : no adhesives, ** : classified, *** : wood free refuse paper

The differentiation within types enables collectors and paper manufacturers to realise technologies that suits the paper to be manufactured and the setting of technical parameters which fit the paper recycling machines much better.

This new standard, complemented with modifications, will fail to give appropriate guidance – mainly because of lack of information – about the quality of refuse paper and the derivable recycled fibres, giving the collectors a difficult task.

In our experimental work we used a significant quantity of the quality "coloured periodicals***", mostly advertisement prints.

The aim of this research work was to find a method for recycling coloured refuse paper in a way that avoids the costly chemical de-inking process for the removal of additives and pigments. The use of coloured refuse paper – as an alternative raw material option – avoids the process of chipping round wood raw material (a very energy intensive technology) since pre-processed paper can be cut into smaller fractions with a properly chosen post-processing technology.

We decided to use coloured periodicals from the category of moderate quality refuse paper (B) because they are used in great quantities and are appropriate for producing board-based, organic composite products after suitable mechanic preparation without adhesives.

2 METHODS

2.1 Materials used, preparation

The selected raw material was refuse paper of almost homogeneous quality. It was coloured, highly glossy paper grade from household wastes. This paper grade consisted of magazines, newspapers, leaflets, catalogues and programme guides. The samples were selected from different, separate containers. The plastic contaminants and metallic buckles were removed by hand after the sorting process. We used the principle of gradualism during preparation following the recycling guidelines of the wood industry.

2.2 Disintegration, determination of fibre dimensions according to ISO 3310

Disintegration was performed using a VIKING GE 110 dis-integrator. The paper fractions were further disintegrated using a Retsch Mühle SK1 equipment, maintaining the length of the fibres. During the disintegration process a conidur sieve was used, which changed the surface of fibres due to the influence of heat and wet state. This surface modification helped the adhesion between the fibres at a later stage. The characterization of the fibres was performed at a fibre moisture content of 8–10% with a particle size analyser. The sampling was a random selection from the pulp. The pulp was classified using 10 different sieves. After the proper vibrating time, the distribution of the fibre fractions was automatically calculated, recorded and displayed.

The parameters for analysis were as follows:

- **A**: vibrating amplitude (mm)
- **T**: vibrating time (min)
- $\mathbf{T_m}$: interval between 2 vibration (s)
- **m**: mass of the initial sample (g)

The parameters for the first trial:

1. A= 1.5 (mm); T= 30 (min); T_m= 10 (s); m= 29.8 (g)

The parameters for the second trial:

2. A= 2.5 (mm); T= 30 (min); T_m= 5 (s); m= 24.4 (g)

The parameters for the third trial:

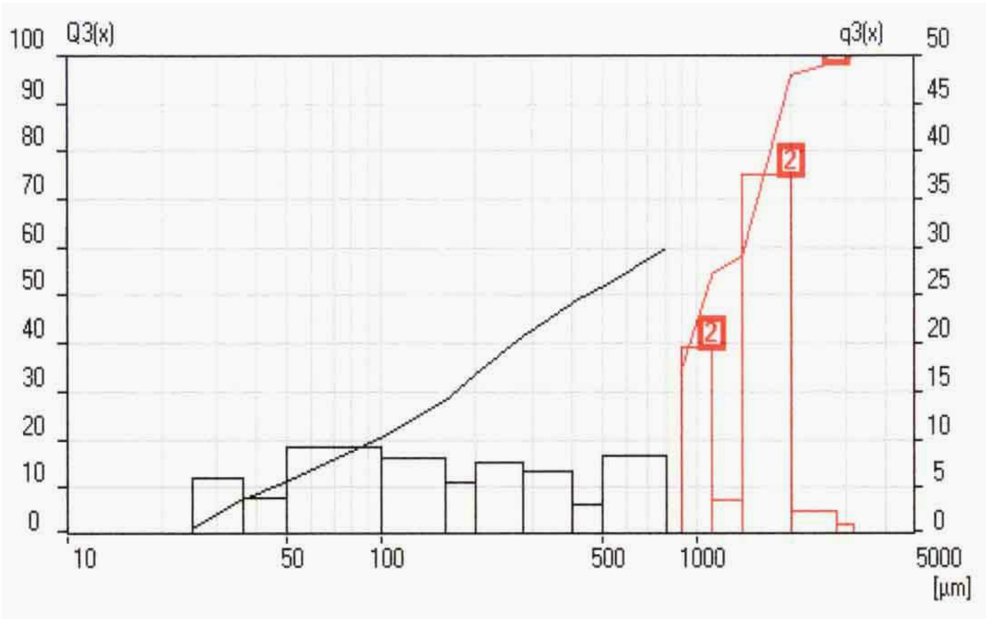
3. A= 2.0 (mm); T= 40 (min); T_m= 5 (s); m= 21.2 (g)

The parameters for the fourth trial:

4. A= 2.5 (mm); T= 40 (min); T_m= 5 (s); m= 25.1 (g)

The properties of the produced panels depend on the disintegration technique, the type of the machine and the produced fibre structure. We could observe that the vibrating amplitude had to be increased from 1.5 to 2.5 mm and we had to shorten the intervals between the vibrations in order to avoid the development of fibre bundles. The fibre fractions showed appropriate differentiation when vibrating time was increased from 30 to 40 minutes.

The results were plotted in columns and solid line format according to the hole diameter of sieves and percentages *(Figure 3)*. In the course of the experiments fibres smaller than 25 μm were present in the sample in insignificant quantities. The fibre fractions were evenly distributed with good approximation. The other fibre fractions were of 36, 50, 100, 160, 200, 280, 400, 500, 800, 1000, 1250, 1400, 1600, 1800, 2240, 2800, 3150, 3550 μm.



Q3(x) – weight distribution sum %
q3(x) – weight distribution density % (marked with "2")

Figure 3. The sample fibre fraction distribution

2.3 Mat preparation, pre-pressing

After disintegration fibres are mixed with adhesives and complementary materials in fibreboard production. There was deliberately no artificial adhesive added to the mat, but it may be used in the near future to improve the properties of the panel. A pre-calculated amount of furnish was then hand-felted into a forming box and pre-pressed into a mat *(Figure 4).*

Figure 4. Single-layer paper fibre mat

The target specific gravity was adjusted using a micro balance. Dry process panels with dimensions of 300x300x16 mm were made from the recycled paper furnish.

2.4 Heat pressing

The volumetric density together with the modulus of elasticity is increasing rapidly during heat pressing. The fibre has visco-elastic behaviour, hence the force-elongation relationship depends on time as well as velocity. Under heat treatment, the natural polymers become plastic and the given volumetric density can be reached by the application of lower pressure. The pressed material springs back less when released. However a board that is kept under constant pressure is creeping.

So heat-pressing is a complex thermo-dynamic process, in which several phenomena take place at the same time and interact with each other. The inner temperature relations during the heat-pressing is very similar to high-temperature drying, water can turn to saturated steam. During the warming phase when the temperature reaches about 100°C, it stays steady while a part of the heat is used for the water to evaporate. The maximum inner pressure has an effect on the strength properties of the panel, primarily on the z-span tensile strength. If the inner pressure reaches 1.0 N/mm^2, after opening the press, a so-called board explosion can occur which decreases z-span tensile strength. Apart from the above-mentioned factors, the geometrical properties of the fibres also determine the quality properties of the produced panel.

Fibre diameter, cell wall thickness, diameter of cell lumen and surface properties of fibres all play important roles in panel quality. The compressability of fibres depends on the ratio of cell lumen and cell wall thickness. Fibres with thin wall cells and bigger cell lumen can collapse more easily. In the case of paper fibres these disadvantageous properties can not be observed. When we increase the fine content, the panels have higher volumetric density and hence higher bending strength and swelling properties. The pressing was done using SIEMPELKAMP 600x600 mm laboratory hot-press equipment, at 185°C on average *(Figure 5).*

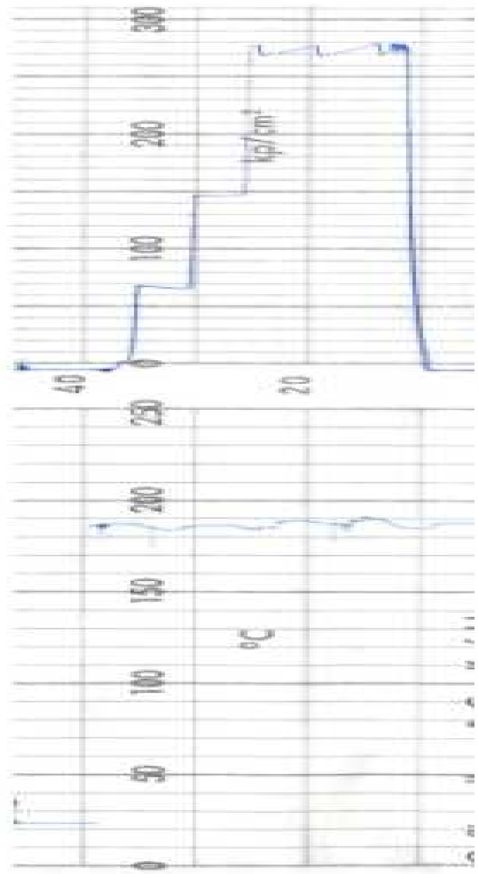


Figure 5. A sample press curve according to temperature and magnitude

2.5 Conditioning

One of the most important procedures of panel manufacturing is conditioning. If the panels are placed on each other after hot pressing they can loose their original strength rapidly. Above 70°C and at relatively high humidity, a hydrolysis effect occurs and results in strength decreasing and bond breaking. In order to avoid this, composite panels need to cool down and let be conditioned, this takes about 4–5 hours at 20°C and at 65% relative humidity.

3 RESULTS

We compared the results of our experiments with the characteristics of standard MDF panels in *Table 2 and 3*.

Table 2. Requirements of fibre panels (EN 622-1-5.)

Name	MDF thickness (mm)				
	<6	6–12	12–19	19–30	>30
Density kg/m^3 (EN 323)	560–900	560–900	560–900	560–900	560–900
Moisture content % (EN 322)	4–11	4–11	4–11	4–11	4–11
Thickness swelling 24 h % (EN 317)	30	15	12	10	8
Modulus of elasticity N/mm^2 (EN 310)	23	22	20	18	17
Internal bond (N/mm)	–	–	–	55–85	–

Table 3. Properties of organic composites

Name		Organic composite
Density kg/m^3 (EN 323)		900 kg/m^3 <
Moisture content % (EN 322)		9 ± 3
Thickness swelling % (EN 317)		80
Bending stiffness N/mm^2 (EN 310)	1000 kg/m^3	8
	1200 kg/m^3	13
	1400 kg/m^3	26
Screw holding N/mm (MSZ 2364)	1000 kg/m^3	52
	1200 kg/m^3	55
	1400 kg/m^3	58

The bending stiffness of the organic composites made without adhesives was similar to that of MDF panels. However, their density was very high.

Screw tests (Dívós 1999) showed good results. They met all the demanded requirements.

Thickness swelling values showed disadvantageous results. Paper fibres are very hygroscopic and a 2-hour-long swelling demolishes the integrity of the panel almost completely.

Still, the application of paper fibres indicate new possibilities in manufacturing fibre panel composite materials.

4 CONCLUSIONS

- Paper fibre, as raw material, has environmentally friendly properties, and the new recycling application we suggest has several potentials.
- This type of coloured refuse paper usage simplifies the recycling techniques used in the paper industry as well as in environmental protection significantly, because this proposed method is a more environmentally friendly than standard paper recycling.
- The raw material supply is simple and cheap, because this type of paper source is available in huge amounts.
- The elaboration and application of this technology can open new possibilities for paper recycling and for lignocellulose-composite materials *(Figure 6).*

Figure 6. From raw material to final product

- The coloured paper waste has visco-elastic properties, hence constant pressing provides better density distribution inside the panel.
- Hot-pressed panels need to cool down gradually and the final moisture content has fundamental impact on the eventual strength of the panels. The cooled down panels were placed on each other for several days in order to avoid later warpage. This condition helps to reach the equilibrium moisture content (9,0%). No adhesives were added to the panels so the originally used complements were activated again and participated in the bonding mechanism besides the fibre-fibre reactions and the secondary chemical bonds between cellulose molecules *(Figure 7–8).*

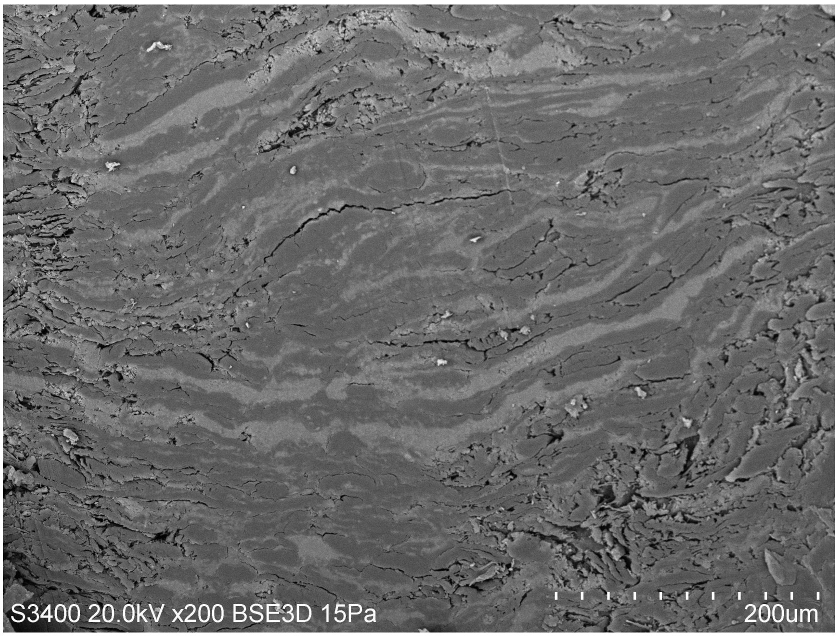


Figure 7. Fibre-fibre bonds in the panel structure
(Amplification: 200 fold)

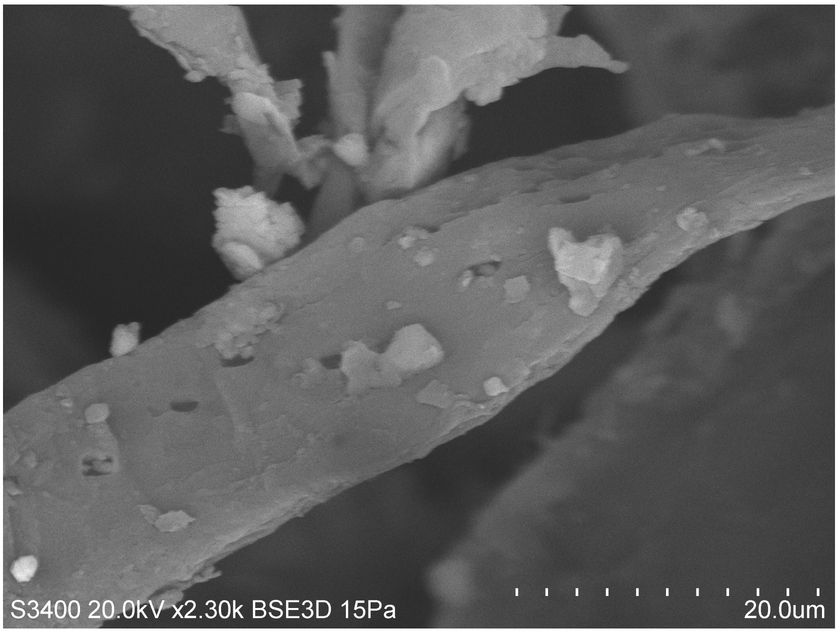


Figure 8. Activated aggregates and starch on the surface of fibre tracheid
(Amplification: 2300 fold)

The obtained results approached the requirements of the MDF panels used in the furniture industry. The cautious application of adhesives can increase some properties, decrease the thickness swelling caused by hygroscopic behaviour and improve the utilization possibilities of this new eco-product.

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