Olli Väntsi

UTILIZATION OF RECYCLED MINERAL WOOL AS FILLER IN WOOD PLASTIC COMPOSITES

Thesis for the degree of Doctor of Science (Technology) to be presented with due permission for public examination and criticism in the Auditorium 1383 at Lappeenranta University of Technology, Lappeenranta, Finland on the 9th of October, 2015, at noon.

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Lappeenrantaensis 663
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Abstract

Olli Väntsi  
Utilization of recycled mineral wool as filler in wood plastic composites  
Lappeenranta 2015  
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The impact of a recycled mineral wool filler on the various properties of wood plastic composites was studied and the critical factors affecting the formation of the properties were determined. An estimation of the volume of mineral wool fiber waste generated in the European Union between the years 2010-2020 was presented. Furthermore, the effect of fiber pre-treatment on the properties of the wood plastic composites were studied, and the environmental performance of a wood plastic composite containing recycled mineral fibers was assessed. The results showed that the volumes of construction and demolition waste and new mineral wool produced in the European Union are growing annually, and therefore also the volumes of recycled mineral wool waste generated are increasing. The study showed that the addition of recycled mineral wool into composites can enhance some of the mechanical properties and increase the moisture resistance properties of the composites notably. Recycled mineral wool as a filler in wood plastic composites can also improve the fire resistance properties of composites, but it does not protect the polymer matrix from pyrolysis. Fiber pre-treatment with silane solution improved some of the mechanical properties, but generally the use of maleated polypropylene as the coupling agent led to better mechanical and moisture resistance properties. The environmental performance of recycled mineral wool as the filler in wood plastic composites was superior compared to glass fibers. According to the findings, recycled mineral wool fibers can provide a technically and environmentally viable alternative to the traditional inorganic filler materials used in wood plastic composites.

Keywords: wood plastic composite, mineral wool, recycling, waste, properties
Acknowledgements

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Olli Väntsi
September 2015
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Author's contribution

In Paper I, the author had the responsibility of choosing the modelling method, collecting the data, and writing the text.

In Paper II-IV the author had the responsibility of sampling, measuring, and analyzing, as well as writing the text.

In Paper V, the author had the responsibility of choosing the assessment method, conducting the assessment, analyzing the results, as well as writing the text.
Abbreviations

1,3-PDO  1,3-propanediol
3D  Three dimensional
ABS  Acrylonitrile butadiene styrene
ADP (fossil)  Abiotic depletion of fossil resources
ADP (element)  Abiotic depletion of element resources
AP  Acidification potential
C&D  Construction & demolition
CB  Carbon black
CCA  Chromated copper arsenate
CCD  Charge-coupled device
CML  The Institute of Environmental Sciences of Leiden University
CR  China reed
EP  Eutrophication potential
EU  European Union
FDM  Fused deposition modelling
GWP  Global warming potential
HRR  Heat release rate
LCA  Life cycle assessment
MAPP  Maleated polypropylene
MC  Moisture content
NIR  Near-infrared
MW  Mineral wool
N/A  Not available
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tr>
<td>NR</td>
<td>Not reported</td>
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<tr>
<td>PA</td>
<td>Polyamide</td>
</tr>
<tr>
<td>PA11</td>
<td>Polyamide 11</td>
</tr>
<tr>
<td>PA 6,66</td>
<td>Polyamide 6/66</td>
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<tr>
<td>PA 610</td>
<td>Polyamide 610</td>
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<tr>
<td>p.a.</td>
<td>per annum</td>
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<tr>
<td>PB</td>
<td>Polybutylene</td>
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<tr>
<td>PBS</td>
<td>Polybutylene succinate</td>
</tr>
<tr>
<td>PBSA</td>
<td>Polybutylene succinate-co-adipate</td>
</tr>
<tr>
<td>PBSAT</td>
<td>Poly(butylene succinate-co-adipate-co-terephthalate)</td>
</tr>
<tr>
<td>PBSL</td>
<td>Poly(butylene succinate-co-L-lactate)</td>
</tr>
<tr>
<td>PBST</td>
<td>Poly(butylene succinate-co-butylene terephthalate)</td>
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<tr>
<td>PBT</td>
<td>Poly(butylene terephthalate)</td>
</tr>
<tr>
<td>PCL</td>
<td>Poly(ε-caprolactone)</td>
</tr>
<tr>
<td>PE</td>
<td>Polyethylene</td>
</tr>
<tr>
<td>PEIT</td>
<td>Polyethylene terephthalate-containing isosorbide</td>
</tr>
<tr>
<td>PET</td>
<td>Polyethylene terephthalate</td>
</tr>
<tr>
<td>PHA</td>
<td>Polyhydroxyalkanoate</td>
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<tr>
<td>PLA</td>
<td>Polylactic acid</td>
</tr>
<tr>
<td>PO3G</td>
<td>Polytrimethylene ether glycol</td>
</tr>
<tr>
<td>PP</td>
<td>Polypropylene</td>
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<td>PTMAT</td>
<td>Polytetramethylene adipate terephthalate</td>
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<td>PTT</td>
<td>Polytrimethylene terephthalate</td>
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<td>PUR</td>
<td>Polyurethane</td>
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<td>Abbreviations</td>
<td>Descriptions</td>
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<td>--------------------------------------------------------</td>
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<tr>
<td>PVC</td>
<td>Polyvinyl chloride</td>
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<tr>
<td>RGB</td>
<td>RGB color space</td>
</tr>
<tr>
<td>REACH</td>
<td>Registration, Evaluation, Authorization and Restriction of Chemicals</td>
</tr>
<tr>
<td>rpm</td>
<td>revolutions per minute</td>
</tr>
<tr>
<td>SBI</td>
<td>Single burning item</td>
</tr>
<tr>
<td>SBR</td>
<td>Styrene-butadiene random (copolymer)</td>
</tr>
<tr>
<td>SEM</td>
<td>Scanning electron microscopy</td>
</tr>
<tr>
<td>THR</td>
<td>Total heat release</td>
</tr>
<tr>
<td>TPS</td>
<td>Thermoplastic starch</td>
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<tr>
<td>UV</td>
<td>Ultraviolet</td>
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<td>WPC</td>
<td>Wood plastic composite</td>
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In the present work, variables and constants are denoted with *slanted style*, vectors are denoted with *bold regular style*, and abbreviations are denoted with regular style.
1 Introduction

1.1 Wood plastic composites

Wood plastic composites (WPCs) are a rapidly developing area of polymer science. Typically WPCs consist of a polymer matrix, wood or other lignocellulosic fibers and additives (Haider et al. 2012). Lignocellulosic fibers have low cost, low density, and high specific properties. They are also biodegradable and non-abrasive (Nabi Saheb & Jog 1999). Coupling agents, lubricants, colorants, flame retardants, and different inorganic fillers are the most common additives in WPCs (Coutinho et al. 1997, Li et al. 2006, Huuhilo et al. 2010b, Stark et al. 2010). WPCs have a wide range of applications, including decking products, automotive parts, and construction products (Clemons 2002, Ashori 2008b, Ndiaye et al. 2008).

The polymer matrix has crucial role in the formation of the final properties of composite products. Traditionally, petroleum-based polymers, such as polypropylene (PP), polyethylene (PE), and epoxy, have been used as a matrix. Lately, polymers from renewable resources have received increasing attention. This development has been mainly due to environmental concerns and the realization that there are finite petroleum resources available. The main drawback of polymers from renewable resources is their degradability during use phase, which can limit the environments where the composites can be used. Other limitations of renewable polymers are their lower softening temperature, which also sets requirements for the end use applications, as well as generally lower mechanical properties compared to petroleum-based polymers (Yu et al. 2006). Some current and emerging plastics are presented in Figure 1.
Improving the interfacial adhesion is especially important in the case of wood and other lignocellulosic fibers, as the unmodified interface between the hydrophilic fibers and hydrophobic polymer matrix has poor quality (Bledzki & Faruk 2003). Coupling agents can improve the strength of interfacial adhesion between the wood fibers and polymer matrix, and also improve the dispersion of the fibers in the matrix, both leading to an improvement in the properties of the resulting composites (Kazayawoko et al. 1999, Lu et al. 2005).
1 Introduction

Lubricants are used to modify the rheology of wood plastic composite melts in the production process. Increasing the output in extruding applications is an example of typical lubricant use. A direct effect of increased material output is a decrease in the overall compounding costs. Like coupling agents, lubricants can also improve the dispersion of filler particles in the polymer matrix. Different stearates, such as zinc stearates, are common lubricating agents in wood plastic composites. Stearates are cost-effective lubricants, but they may not be compatible with maleated coupling agents. In such cases, other lubricating agents, for example, amide- or ester-based agents could be viable. (Markarian 2002, Satov 2008)

Colorants are used in WPCs for aesthetic reasons and ultraviolet (UV) protection. To achieve consistent color, it may be necessary to add the colorant to both the wood fibers and the polymer matrix separately prior compounding. Pigments, especially ones with dark color, can be used to reduce the discoloration of WPCs in weathering. However, they are not able to stop the chemical changes occurring in the wood fibers and polymer matrix caused by weathering. (Rowell 2007, Butylina et al. 2012)

WPCs have many applications, such as various construction applications where good reaction-to-fire properties are needed. Magnesium hydroxide, boric acid, ammonium phosphates and borates, ammonium sulfate and chloride, zinc chloride and borate, phosphoric acid, dicyanodiamide, sodium borate, and antimony oxide are common fire retardant agents used in wood plastic composites. In addition, some inert filler materials, such as glass fibers or talc can improve the fire performance of WPCs. (Nikolaeva & Kärki 2011)

Inorganic fillers for wood polymer composites include, for example, calcium carbonate, wollastonite, soapstone, talc, nanoclay, silica, and glass fibers. These inorganic fillers have shown potential in improving the mechanical, fire retardant and thermal properties of WPCs (Faruk & Matuana 2008, Rizvi & Semeralul 2008, Huuhilo et al. 2010b, Deka & Maji 2012). Inorganic fillers are also cheaper than polymers, and therefore, the raw material costs of WPCs can decrease when polymers are replaced with inorganic fillers (Klyosov 2007).

1.1.1 Processing of wood plastic composites

Extrusion, injection molding, compression molding and additive manufacturing technologies can be used to process WPCs. Extrusion and injection molding are the most common WPC processing technologies, while additive manufacturing technologies, such as 3D printing of WPC are strongly emerging processing solutions. (Kim & Pal 2010, Faruk et al. 2012, Hofmann 2014)

In the extrusion process the polymer is melted and wood and other additives are then mixed into the melted polymer. The compounded mixture is then conveyed through a die to give the product the desired shape. Hollow and solid profiles, granulates, and other semi-finished products are examples of typical products of the extrusion process. Single
screw extruders, counter-rotating twin screw extruders and co-rotating twin screw extruders are common types of extrusion machines. Twin screw extruders are often used in large-scale applications because they provide better dispersion of the melt and higher material output compared to single screw extruders. The counter-rotating twin screw extruder has lower screw revolutions per minute (rpm), which reduces the risk of burning the materials, as well as lower shear mixing compared to the co-rotating twin screw extruder. The advantage of the co-rotating twin screw extruder is the ability to process materials at ambient moisture content, as the extruder is capable of drying the materials, unlike counter-rotating extruders with which additional drying equipment is needed (Kim & Pal 2010, Faruk et al. 2012). Extrusion is a suitable processing method for polymers with high molecular weight, as high molecular weight provides better melt strength (Faruk et al. 2012).

Injection molding is a widely used technique in the wood plastic composite industry. Injection molding makes it possible to produce products with complex geometries. Injection molding is also a fast and economically effective production process suitable for large scale production. Injection-molded products require little to none finishing. Polymers with low molecular weight, and therefore, low viscosity, are best suited for injection molding (Faruk et al. 2012). A schematic view of injection molding is presented in Figure 2.

![Schematic view of injection molding](The Open University 2010)

While additive manufacturing technologies, such as stereolithography or selective laser sintering, have been commercially available since the 1980’s, additive manufacturing technologies have received more attention in the WPC industry only recently. Especially, the development of fused deposition modeling (FDM) has brought the prices and technological feasibility of additive manufacturing technologies to an attractive level. In the FDM concept the plastic material, possibly containing wood and/or other additives,
is extruded through a thin heated nozzle. The nozzle movement can be controlled in the x-y-z-direction, which allows rapid manufacturing of products with complex geometries. The main advantage of FDM is its ability to produce unique products rapidly and very cost effectively, for example, prototyping and personalized medical prostheses are very attractive applications for FDM. In mass production, the other production processes, such as injection molding, are currently more effective. The polymers used in FDM are typically polylactic acid (PLA) and acrylonitrile butadiene styrene (ABS), but the use of polycarbonate, polyamide or polyphenyl sulfone is possible, especially with higher-grade machines. (Hofmann 2014)

1.1.2 Development of the wood plastic composite market

The increased environmental awareness of consumers, as well as attempts to reduce the dependency on non-renewable resources, such as oil, have acted as drivers for the development of the WPC market. North America has the largest share of WPC markets, estimated to be 48% of the total markets in 2015. China is the second largest market with the share of 33%, followed by Europe with a 9% share and Japan with a 4% market share. In relative growth of the market, China is clearly the fastest growing market, with an annual growth of 25%. The growth per annum (p.a.) in North America is 8% and 11% in Europe (Eder & Carus 2013). According to forecasts, the total market in the European Union is expected to rise from 265 000 tonnes p.a. in 2012 to 580 000-950 000 tonnes p.a. in 2020, depending on the incentives for bio-based products (Carus et al. 2014).

In Europe, decking products dominate the WPC market; their share of the total WPC market was 67% in 2012. Products for the automotive industry were the second largest group, with the market share of 23%. Siding and fencing products, technical applications, and furniture are other notable product areas in Europe. Automotive applications are expected to present the largest relative growth, as their share is estimated to raise from 60 000 tonnes in 2012 to 80 000-300 000 tonnes in 2020. (Carus et al. 2014)

Market development strategies to increase the market share of companies producing products from recycled materials include proper advertising based on the good price-quality-ratio of a recycled material or product, environmental performance, and material efficiency. The emphasis of advertising can vary depending on the product and company. Companies using virgin raw materials often put emphasis in their advertising either on high quality and high reliability or on a combination these properties. (Batzias & Pollalis 2009)

A company marketing recycled products must find a delicate balance between environmental, functional, and economic values. Over-exaggeration of environmental values may create a negative customer attitude towards the product or company, especially, if consumers feel that the environmental claims are misleading or outright false. For this reason, it is important that the environmental claims are based on straightforward evidence, and marketing communication is kept simple enough to
minimize the possibilities of the marketing message being misinterpreted by consumers. (D'Souza et al. 2006)

It is clear that the environmental performance of materials and products are becoming increasingly important in the development of market shares (Akehurst et al. 2012). The utilization of recycled materials in WPCs can offer many possible market advantages: improved environmental performance of the product, improved material efficiency, and good price/quality-ratio. It must be ensured that the marketing of such products is done according to good practices in order to avoid negative consumer attitudes towards a product or even towards a whole product category. Considering the market development of wood plastic composites and environmentally efficient products in general, there is great market potential in wood plastic composites utilizing recycled raw materials.

1.2 Recycled raw materials in composites

WPCs provide many options for the utilization of recycled raw materials. Recycled wood fibers and polymer materials are used in many commercial grade products (Ashori 2008a). The use of waste materials can help to minimize the amount of waste disposed at landfills, which will reduce the environmental emissions originating from landfill deposition. Furthermore, natural resources are conserved when the need for virgin raw materials is reduced (Weber et al. 2002, Poon 2007).

The use of recycled polymers in WPCs has been studied extensively, and recycled polymers are commercially available from vendors. Recycled polymers can be used to produce WPCs with mechanical and moisture resistance properties comparable to WPCs made of virgin polymers (Adhikary et al. 2008, Ashori & Nourbakhsh 2009). While various recycled polymers are commercial, many studies also describe the utilization of recycled polymers from non-commercial sources. Ashori (2008a) mentions municipal solid waste as a potential source of recycled plastic for WPC production. Lei and Wu (2011) have used recycled oil containers to prepare WPCs. Jayaraman and Bhattacharyya (2004) obtained recycled plastics from a kerbside recycling scheme to produce WPCs. Both sorted and mixed plastic fractions were used in the study. Singleton et al. (2003) used recycled polyethylene bottles as the matrix in WPCs. In all these studies, the authors found recycled plastic very promising raw material for WPC production.

The wood or other lignocellulosic fibers used in wood plastic composites are often byproducts of industrial or agricultural processes. Najafi et al. (2006) used sawdust, which is a common example of an industrial byproduct, in the production of wood plastic composites. Migneault et al. (2014) compared the feasibility of natural fibers from different sources and found that sawdust can provide functional properties comparable to virgin wood fibers in wood plastic composites. The cellulose content of the fiber is one of the main factors explaining the differences in the functional properties of WPCs, and there was no significant difference in the cellulose content of virgin wood fibers and sawdust.
Recycled newspaper and paper sludge are also possible sources of natural fibers for wood plastic composite production. Recycled newspaper, when used together with a coupling agent, provides good mechanical properties for composites. The properties remained feasible even with very high (85%) recycled newspaper content according to Ashori and Nourbakhsh (2009). Sanadi et al. (1994) name dispersion of the fibers and the interfacial adhesion of the fibers and polymer matrix as the main factors affecting the mechanical properties of recycled newspaper-filled polymers. Both these factors can be improved with the proper use of a coupling agent. Furthermore, a high energy kinetic mixer can help to disperse the newspaper fibers in the matrix better. Huda et al. (2005) note that recycled newspaper tends to have high cellulosic content, around 75%, with the rest of the fiber composed of ash, which is a combination of the carbon left after burning and any organic and non-organic substances that are not volatilized after the heating of the cellulose fiber in the production phase. It has been shown that α-cellulose fibers, which are fibers from which lignin and hemicellulose have been removed, improve the properties of WPCs compared to wood fibers (Ou et al. 2014).

Corn stover, coir, bagasse and wheat or rice straws are agricultural byproducts which can be used to produce lignocellulosic fibers suitable for composite production. The chemical composition of agricultural fibers can differ quite a lot from each other, and therefore, it is important to choose the fiber suited best for each different application. It also possible to modify the agricultural fibers to improve their usability. Low prices and good availability are the main advantages of agricultural byproducts. (Reddy & Yang 2005)

The different additives in WPCs are often virgin materials, although some recycled inorganic fillers, mainly glass fibers, have been studied. Valente et al. (2011) compared virgin and recycled glass fibers as fillers in WPCs. The recycled glass fibers were obtained from mechanical recycling of glass fiber-reinforced polyester composites. They report a slight decrease in the flexural strength and screw withdrawal resistance of composites containing recycled glass fibers compared to composites containing virgin glass fibers. Zheng et al. (2010) recovered recycled glass fibers from the nonmetals of waste printed circuit boards. Using recycled glass fibers as the filler in PP clearly improved the mechanical properties compared to clean PP.

1.3 Construction waste in Europe

1.3.1 General approach

The generation of construction waste in the 27 countries of the European Union (EU27) is presented in Figure 3, which includes the waste generated by all activities related to the construction industry. Construction-related waste accounted for 30.0-35.6% of the total waste generated in the EU27 area during the period 2004-2012 (Eurostat 2015).
While the construction industry is a major source of waste, the composition of the waste stream originating from construction activities is not precisely known (Llatas 2011). The European Union provides a list of wastes as a guideline for classification, but such accuracy is only rarely implemented in actual studies regarding the composition of construction-related waste. The Eurostat database lists the breakdown of construction-related waste according to the classification guidelines on the Member State level, which can cause inaccuracies due to different levels of control and reporting of construction-related waste in the Member States (Sáez et al. 2011).

The wood and plastic fractions are commonly listed separately in case studies concerning the composition of construction-related waste. Furthermore, metals are often presented as their own category, but the metal fraction is less relevant from the wood plastic composite point of view. Mineral wastes are commonly reported as one category, but very heterogeneous material composition can be included in this category (Bergsdal et al. 2007, Lau et al. 2008, Del Río Merino et al. 2010). It is not possible to differentiate mineral wool fibers from other mineral waste due to lack of data, even though the waste list of the European Union has a separate classification code for non-asbestos-containing insulation materials (European Commission 2000).

1.3.2 Properties of mineral wool obtainable in the construction waste stream

The term mineral wool covers both rock wool, produced from volcanic rock, such as diabase or basalt, together with limestone and/or dolomite, and glass wool, produced from quartz, soda and limestone. Recycled glass is also often used in the production of glass wool. Mineral wool fibers can be bound together by adhesives, typically phenol-formaldehyde resin, in certain insulation products. The binder can account for 10% of the total mass of the finished product. Mineral or silicone oils are used as lubricants in
the mineral wool production process, they usually amount up to 1% of the final product mass. Mineral wool can be laminated with aluminum foil, kraft paper or non-woven glass fiber mats. (Müller et al. 2009)

The chemical composition of mineral wool depends on the raw materials used, and is slightly different for rock wool and glass wool. Some examples of the chemical composition of mineral wool are listed in Table 1. The fiber thickness of new mineral wool is normally between 5 and 10 µm and the thermal conductivity between 0.035-0.045 W/(m·K) (Širok et al. 2008, Müller et al. 2009). Sal'nikov (2003) has investigated the properties of mineral wool after 40-50 years of service life and found that neither the fiber diameter nor the thermal conductivity differed notably from newly manufactured mineral wool.

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<tbody>
<tr>
<td>SiO₂</td>
<td>46.43</td>
<td>40-52</td>
<td>56.89</td>
<td>39.61</td>
<td>45.32</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>11.42</td>
<td>8-13</td>
<td>3.47</td>
<td>16.68</td>
<td>15.61</td>
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<tr>
<td>TiO₂</td>
<td>1.47</td>
<td>1.5-2.7</td>
<td>0.12</td>
<td>NR</td>
<td>NR</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>4.41</td>
<td>5.5-6.5</td>
<td>0.57</td>
<td>1.39</td>
<td>10.15</td>
</tr>
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<td>FeO</td>
<td>4.72</td>
<td>NR</td>
<td>0.18</td>
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<tr>
<td>MnO</td>
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<td>0.56</td>
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<td>CaO</td>
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<td>10-12</td>
<td>12.61</td>
<td>37.30</td>
<td>20.72</td>
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<td>MgO</td>
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<td>8-15</td>
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<td>6.45</td>
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<tr>
<td>Na₂O</td>
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<tr>
<td>B₂O₃</td>
<td>0</td>
<td>NR</td>
<td>6</td>
<td>NR</td>
<td>NR</td>
</tr>
</tbody>
</table>

*NR=not reported*

Both rock wool and glass wool are considered noncombustible; the building material class for rock wool is A1 or A2, depending on the product, and A2 for glass wool. Recycled mineral wool can contain various contaminants, such as secondary components used in the manufacturing, which must be taken into consideration when evaluating the re-use potential. The changes over the service life of mineral wool must also be considered: foreign components used in the construction industry, such as fasteners, reinforcement fabrics, foils, plaster materials, or sealing tapes can easily get mixed into the mineral wool waste (Müller et al. 2009). Possible fungal or bacterial growth occurring during the service life of mineral wool, especially in moisture-damaged buildings, must be taken into consideration (Hyvärinen et al. 2002).
1.4 Drivers and barriers for recycled material utilization

1.4.1 Drivers

Environmental laws and regulations are one of the most important factors promoting the use of recycled raw materials, not only with WPCs, but in all industries. In the European Union, the waste framework directive (directive 2008/98/EC) states that the Member States shall take the necessary measures designed to achieve that by 2020, preparing for re-use, recycling and other material recovery, including backfilling operations using waste to substitute other materials, of non-hazardous C&D waste, excluding certain naturally occurring materials, shall be increased to a minimum of 70 % by weight (European Commission 2008).

The waste hierarchy for the European Union is also introduced in the waste framework directive. It divides the waste prevention and treatment methods into five different categories, as shown in Figure 4. The prevention of the waste should be the waste management option of highest priority. Effective waste management strategies are seen as the main tools in waste prevention. (Formoso et al. 2002)

Preparing for re-use is the second highest priority, it is defined in the waste framework directive as “checking, cleaning or repairing recovery operations, by which products or components of products that have become waste are prepared so that they can be re-used without any other pre-processing” (European Commission 2008). For example, injecting fine powdered waste materials back to the melting furnace in stone wool production can be considered as preparing for re-use, as the material is used in the same process it originated from and no processing of the recycled material is needed. (Karppinen 2004)
Recycling is defined as reprocessing recycled material into products, materials or substances to be used in the original or other purposes. Energy recovery or reprocessing recycled material into fuels are not considered recycling (European Commission 2008). Examples of recycling are much more common than preparing for re-use. Mineral wool waste can be processed into briquettes which can be used as raw materials in the manufacturing of mineral wool, the difference to direct injection into the melting furnace is the need to process the waste material (Balkevičius et al. 2007). Processing mineral wool waste into raw materials for other products, such as cement-based composites, wood-mineral wool hybrid particleboards or horticultural products, is also considered recycling (Grunert et al. 2008, Cheng et al. 2011, Mamiński et al. 2011).

The second last option, recovery, is defined as recovery methods other than the ones introduced in the higher categories. Energy recovery and processing waste materials into fuels are included in this category (European Commission 2008). As non-combustible material, recycled mineral wool has little potential in this category.

The least preferred option of waste treatment is disposal. Disposal means operations where waste material is discarded without any recovery. Landfill deposition is the most common disposal method, but all activities where waste material is disposed into the nature are also considered to belong to this category. The incineration of waste material without energy recovery is also included in the disposal category (European Commission 2008). With the lack of widely available recycling options, mineral wool waste is often disposed at landfill.
The environmental consciousness of consumers has been growing recently, and the environmental performance of products has gained importance in marketing. However, many environmentally friendly products have not achieved the market success which might be expected with the increased environmental consciousness. It can be said that while the environmental consciousness of consumers is evident, ecologically conscious decision-making is not (Wong et al. 1996). A majority of consumers still consider functional and economic performance of the product more important when making their purchasing decisions (Chen et al. 2012). However, the environmental performance of products is still seen as an important factor in both private and public sectors, and therefore should be taken into consideration when designing a product (Giudice et al. 2005). Lastly, the optimization of the environmental performance of the product does not have to mean a compromise in its functional and economic performance, but balancing all these factors is needed for commercially successful sustainable design (Chen et al. 2012).

Effective utilization of recycled materials can provide many environmental benefits. When reduced amounts of waste are deposited to landfills, the environmental effects related to landfills, most importantly different leachates and land use, are also reduced (Weber et al. 2002, Poon 2007). The scarcity of non-regenerative natural resources can also act as a driver of recycled material use. Recycled materials can offer reductions in the consumption of oil, mineral resources and energy, all which are strong incentives for recycling (Siddique et al. 2008). Economic incentives are an important driver in materials recycling. It is possible to achieve economic advantages using recycling materials, but this must usually be evaluated case by case due to different conditions (Marzouk & Azab 2014).

1.4.2 Barriers

Obviously, many barriers to the utilization of recycled materials exist. The quality of recycled material is one of the main concerns. Many manufacturing processes require clean and uncontaminated raw materials (Dunster 2007a). The exact origin of recycled materials is usually not known, and therefore, the material may have been subjected to degradation or contamination during its previous use. The unknown background of the material can lead to manufacturers’ mistrust of the functional performance of recycled material, hindering the large-scale use potential (Vilaplana & Karlsson 2008). A quality control concept to assess the properties of recycled material with defined tolerances could be introduced to satisfy the requirements of manufacturers and customers, guaranteeing the performance of recycled material in their applications (Karlsson 2004).

The safety of recycled materials must be ensured both in the manufacturing and use phase. The safety risks may be targeted at humans or the environment (Guo et al. 2009). Recycled materials may contain hazardous substances, which can be released into the environment during the pre-processing, manufacturing or use phase of the material. Methods to monitor and control the potential toxicity of products made from recycled materials have been developed for certain products, and similar principles could be
adopted and developed further to fit the needs of other production systems (Binderup et al. 2002).

In the case of recycled mineral wool, the main hazardous contaminant can be asbestos, which was previously widely used in construction. Airborne asbestos fibers are associated with an increased risk of lung cancer and mesothelioma (Stayner et al. 1996). It must be ensured that no asbestos is among the recycled mineral wool material. The binders used in the production of mineral wool contain phenol and formaldehyde, both of which can have serious negative health effects, but modern phenol-formaldehyde resins do not release phenol or formaldehyde in amounts which exceed the safety limits given by the Finnish Institute of Occupational Health (Balcerowiak et al. 1995, Mäkinen et al. 1999, Finnish Institute of Occupational Health 2014a,b). Mineral wool fibers have typically relatively low bacterial and fungal growth on them, and it is unlikely that bacteria or fungal concentrations typically found in recovered mineral wool material would pose a health risk during normal recycling, manufacturing and end user operations (Pitt 1994, Hyvärinen et al. 2002).

The hazards associated with mineral wool itself should also be comprehended during the recycling and manufacturing processes. Concerns about the possible health effects of inhalation of mineral wool fibers have been raised. Mineral wool materials can release respirable fibers into the air during normal handling. Due to these concerns, a considerable amount of research has been done to evaluate the health effects of respirable mineral wool fibers (De Vuyst et al. 1995, Kamstrup et al. 1998, Hesterberg & Hart 2001, Kudo & Aizawa 2011). The International Agency for Research on Cancer originally classified mineral wool as “possibly carcinogenic to humans”, but after a reevaluation in 2002 classified them as “not classifiable as to their carcinogenity to humans” (Andersen et al. 2002, Baan & Grosse 2004). The Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) regulation of the European Union classifies mineral wool fibers with diameter <6 µm and soluble component index >18 % by weight as hazard class Carc. 2, which means that they are suspected of causing cancer. The soluble component index is equal to the sum of the percentage by weight composition of alkaline and alkali–earth oxides (Na₂O + K₂O + CaO + MgO + BaO) (European Chemicals Agency 2013). The research done on the subject, despite the various classifications, has not provided clear evidence of the relation between exposure to rock or glass wool and lung cancer or any respiratory diseases (De Vuyst et al. 1995, Hesterberg & Hart 2001). The hazard classification does not necessarily correlate with the actual risk of harm. In some cases the risk of harm from hazardous substances can be reduced to a negligible level by proper use, storage and handling processes. Protective measures, such as personal protection equipment, can also reduce the risk of harm (Bernstein 2007).

Certain economy-related barriers also exist. The direct costs of recycling are often exceed the benefits (Tonjes & Mallikarjun 2013). Costs can occur in the transportation or handling of recycled material. Transportation costs can be responsible for a notable amount of the total costs of a recycling project (Kang & Schoenung 2006). They are especially relevant in the case of mineral wool recycling. Waste mineral wool is bulky
material, and it may be necessary to compress or bale it in order to achieve economically sustainable transportation. Additional investments may be required from companies handling or utilizing recycled mineral wool, as it must be stored and often pre-processed prior to utilization. If suitable infrastructure is not available at the facilities, it may be necessary to invest in building them. (Dunster 2007a).

1.5 Waste separation technologies

C&D waste is generated by three different activities: construction, renovation, and demolition. The waste volumes generated in the activities differ greatly from each other (Wang et al. 2004). Renovation and especially demolition produce much larger waste volumes compared to construction. The C&D waste generated in new construction typically amount for 10-32 % of the total C&D waste generated (Bossink & Brouwers 1996, Coelho & De Brito 2011). Mineral wool waste volumes originating from new construction can be lower, around 5 % of the total mineral wool waste volume, as the left-over mineral wool material can be re-used in other jobs, or in some cases even returned to the manufacturer if a take-back scheme is offered (Dunster 2007b, Müller et al. 2009). It is relatively easy to re-use or recycle the mineral wool waste originating from new construction, as the material composition is known, the material is not necessarily contaminated and it is easy to separate mineral wool waste from other C&D waste fractions manually if needed.

Recovering materials from the renovation and demolition waste stream is a much more complicated issue. These waste streams are often heterogeneous, and recyclable materials must be separated from them (Mulder et al. 2007). In general, two different approaches to waste sorting can be taken: on-site recycling and off-site recycling. On-site recycling is done at the construction site and off-site recycling in a specific waste recycling facility. Both approaches have their strengths and weaknesses.

1.5.1 On-site recycling

The effectiveness of on-site sorting depends on the selected waste management method. The selective demolition method combined with the top-down method offers a relatively easy way to recover recyclable materials (Poon et al. 2001). The selective demolition method can also be applied to renovation solutions (Cha et al. 2012).

Selective demolition is a method where demolition is essentially carried out as the reverse of the construction progress, and recyclable materials are separated from the waste stream (Poon et al. 2001, Kourmpanis et al. 2008). In the top-down method a building is demolished floor by floor from the roof to the ground level using light tools instead of rougher demolition methods, such as explosives or a wrecking ball. The combination of the selective demolition and top-down methods can produce a well-separated and uncontaminated waste stream, but is considerably labor- and time-consuming. All on-site sorting operations require suitable space on the construction site and education of the workforce regarding waste management issues (Poon et al. 2001).
1 Introduction

Wang et al. (2010) have studied the critical success factors of on-site recycling of C&D waste. Manpower was seen as the most critical factor affecting on-site recycling. Extra manpower is needed to recover valuable materials from the waste stream. Furthermore, an operative who is in charge of the material recovery operations should be pointed out to optimize the efficiency of material recovery. Contractors are rarely willing to invest in such practice if they are unable to gain additional benefits by doing so. In order to construction contractors to be able to realize the additional benefits from on-site waste separation, existing markets for recycled materials are required. The slow development of such markets is a factor hindering all C&D waste recycling (Meinander et al. 2012, Mansikkasalo et al. 2014).

Waste sortability has been identified as a critical factor in on-site recycling. The main advantage of on-site recycling compared to off-site recycling is the possibility to separate the waste material fractions before they are mixed, thus removing the need to identify specific materials from the mixed waste stream. The technical implementation of on-site waste separation is a more complicated issue. The technical solutions to keep the material fractions chosen for recovery separated from other waste fractions can be inconvenient to operate. Proposed solutions include separate containers for each fraction chosen for recovery, and possibly a centralized collection system, for example, a chute-based system, to direct the material flows in the containers. (Wang et al. 2010, Yuan et al. 2013)

The management of a construction project has a notable effect on the efficiency of waste recycling. First, good management practices can reduce the overall waste volumes. Clearly defined responsibilities in new construction operations reduce the need to do rework, which again reduces the generated waste volumes. However, reducing the need to do rework is mainly a cost factor, and thus managers are relatively well aware of the need to optimize material usage in new construction projects. The waste management strategies of renovation and demolition do not have as direct cost incentives associated with them. Commonly, with the lack of an economic incentive to recycle, the level of environmental awareness of management and construction operators becomes the main factor influencing waste management decisions. (Wang et al. 2010, Mendis et al. 2013)

Lastly, factors such as site space can be seen as critical factors influencing on-site recycling. The importance of site space depends on the type of construction project, as some projects have more readily available site space. The site space for waste sorting should be included in the layout plan of the construction site. Temporary placement of sorting facilities can cause disruptions in other construction activities. (Poon et al. 2001, Wang et al. 2010)

1.5.2 Off-site recycling

Off-site recycling relies on technological solutions for recovering determined waste fractions from the mixed waste stream in a specific recycling facility. The technological level of off-site facilities varies from manual handpicking to advanced sensor-based automation systems. The composition of the recovered material fraction at off-site
facilities can vary greatly, depending on the technological level of the sorting equipment. In some cases relatively pure material fractions (glass, wood, etc.) can be separated, and in other cases the mixed waste can be separated into two fractions: inert and non-inert material. Possible material use applications of the recovered fractions are largely determined by the quality of the sorting process. (Kourmpanis et al. 2008, Lu & Yuan 2012)

Manual sorting is still widely used in the recycling facilities around the world. It is slow and labor-intensive technology, but can produce relatively homogeneous material fractions (Kourmpanis et al. 2008, Lu & Yuan 2012). However, health risks are associated with manual waste separation. The workers in manual waste separation can be exposed to various hazardous substances. Certain substances can be released into air during waste handling processes and end up being respired. The observed occupational health problems among waste sorting workers include pulmonary diseases, gastrointestinal problems, eye inflammation, irritation of the skin, and upper airway problems (Poulsen et al. 1995).

A number of technological equipment can be used for C&D waste separation. Some of the equipment aim at processing the waste into a form better suited for separation, for example by means of size reduction, while others are focused on actual separation of certain fractions from the waste stream. Size reduction equipment include such techniques as different shredders, hammermills, jaw crushers, and hydraulic breakers that can be used to reduce the size of C&D waste (Peng et al. 1997). Size reduction is usually a rather simple task and the equipment for it is readily available. The actual sorting operations are much more complicated.

The problem in sorting operations is not so much the availability of techniques. Magnetic separators can be used to separate metallic materials from the waste stream. Gravity separators, float tanks and air classifiers can separate materials based on their weight or density. Different sieving equipment can be used to separate different size fractions from each other. In an off-site facility, the waste stream usually passes through multiple techniques. A typical process flowchart of an off-site facility is presented in Figure 5. (Peng et al. 1997)
Figure 5. Process flow chart of a typical off-site sorting facility. (Peng et al. 1997)

The problem is in the limitations of the technologies, as their ability to classify material fractions is limited to the single property or specific properties the apparatus is designed to measure. A float tank cannot tell apart two different material fractions with the same...
density, or a sieve cannot separate two same sized particles from each other. They often do not produce a material fraction pure enough to satisfy the requirements of the markets. Therefore, more advanced technologies are needed to improve the quality of the recovered material fractions. (Vegas et al. 2015)

Advanced sorting technologies are being rapidly developed. They include such technologies as optic systems, near infrared (NIR) sensors, electromagnetic sensors, laser-induced breakdown spectroscopy, and different X-ray sensors. (Linß et al. 2012, Meinander et al. 2012)

Optic systems typically include a charged couple device (CCD) camera, an RGB adapter, a frame grabber, and a computing unit. They have potential uses especially in plastic separation, where the machine vision systems can sort the materials based on their color and opacity. The challenges of the technology are related to the interpretation of the acquired image. It may be challenging to separate one object from another if they overlap in the image. Furthermore, if the material has dirt or other material, such as label stickers, attached to it, the color of the object may be determined incorrectly. (Tachwali et al. 2007)

In many cases, color and opacity information alone is not enough to separate different materials from each other correctly. Optic systems can be combined with NIR systems to produce information about the chemical composition of the object (Vegas et al. 2015). NIR systems are able to obtain the spectral signature of the material and by analyzing these signatures, it is possible to separate the different material fractions from the waste stream more accurately. For example, it is possible to distinguish different plastic materials (PP from PE, etc.) with good accuracy. Optic system should be included in the system, however, as black or dark colored materials are challenging for NIR systems due to their low reflectance (Serranti et al. 2012).

Different X-ray technologies can be used, for example, to differentiate inert materials from each other, to identify polymers, and to separate treated wood from clean wood wastes. The sorting of small size inert material fractions is often challenging, but with X-ray technologies it is possible to separate minerals with specified chemical compositions. For example, it is possible to obtain the mineral fraction directly re-usable in production of concrete from mixed inert waste by using X-ray technology (Bianchini et al. 2005). X-ray technology has been found to be an effective method to separate contaminated and treated wood from the clean wood fraction. Especially chromate copper arsenate (CCA)-treated wood was successfully separated from other wood materials with high accuracy in a study by Fellin et al. (2014). It is possible to identify different types and grades of plastic, also dark colored, using X-ray-based systems (Bezati et al. 2010).

An electromagnetic sorting device can be used to differentiate materials on the basis of their conductivity. Electromagnetic systems can be used, for example, to separate ferrous metals from non-ferrous ones with good accuracy (Brojboiu & Mandache 2011). Laser-induced breakdown spectroscopy can be used to sort different plastic types from the waste.
stream or to identify treated wood from clean wood, much like with the X-ray based systems (Moskal & Hahn 2002, Boueri et al. 2011).

The emerging technologies have great potential in increasing the quality, cost effectiveness and material output of off-site recycling facilities (Vegas et al. 2015). They do not necessarily render traditional sorting technologies useless, but provide increase in the overall efficiency of sorting when used together with traditional methods.

1.6 Life cycle assessment of wood plastic composites

Wood plastic composites are often described as environmentally friendly materials, which, compared to traditional polymer composites, offer environmental advantages such as reduced dependence on non-renewable energy and material sources, lower pollutant emissions, lower greenhouse gas emissions, enhanced energy recovery, and end-of-life biodegradability of some components. (Joshi et al. 2004)

Environmental performance is an important driver of increased future use of WPCs, and it is important to be able to assess the environmental impacts of the composites properly. Life cycle assessment (LCA) is a technique developed to address the environmental aspects and potential environmental impacts (e.g. use of resources and environmental consequences of releases) throughout a product’s life cycle from raw material acquisition through production, use, end-of-life treatment, recycling, and final disposal (i.e. cradle to grave). (Finnish Standards Association 2006)

A list of factors has to be considered before a material can be declared eco-friendly. In the case of polymer composites, things such as the polymer matrix, different additives and filler materials can affect their environmental impacts. The environmental impacts of lignocellulosic fibers have to be considered as well, as the process chain of cultivating, harvesting, and processing the fibers to suit composite production can have a significant impact on the environmental performance of the composites (Corbière-Nicollier et al. 2001, Simões et al. 2012). Figure 6 shows typical processes included in the LCA of lignocellulosic composites.
The environmental impacts of certain lignocellulosic composites have been reported in previous studies. Xu et al. (2008) studied wood polypropylene composites and found that the polymer matrix had a major effect on the environmental impacts of the composite, while the 30 % wood fiber loading had a relatively small impact. It was also found that the transportation of the materials did not have a significant role in the formation of the environmental performance of the composite.

Joshi et al. (2004) studied the environmental properties of glass fiber and natural fiber-filled polymer composites. Natural fibers were found to have superior environmental properties, mainly because the production of natural fibers resulted in lower environmental impacts compared to the production of glass fibers. Natural fiber composites also had higher fiber content for equal functional performance, reducing the amount of a more polluting base polymer. Natural fibers have lower weight compared to glass fibers, which leads to lower weight of natural fiber composites compared to glass fiber composites with equal functional performance. The lower weight can help to reduce environmental impacts in the use phase, for example in automotive applications, and improve the efficiency of the end-of-life options, such as incineration for energy.

Corbière-Nicollier et al. (2001) have also studied the replacement of glass fibers in polymer composites with natural fibers. The fibers used were China reed (CR). They found that the CR fiber composites had improved environmental performance compared to glass fiber composites, as long as the service life of the CR fiber composite is long enough. A service life of 2.2 years, compared to the 5-year service life of glass fiber

Figure 6. Life cycle of a natural fiber composite. (Joshi et al. 2004)
composites was enough to achieve reduced environmental impacts. The recycling rate of the composites also affected their environmental performance, but glass fiber composites would require extremely great advantage in the recycling rate compared to the CR fiber composites in order to achieve better environmental performance. It was also shown that using CR fibers as a raw material had higher substitution potential than the use of CR fibers for heat production or the production of biofuels.

Rajendran et al. (2012) compared the environmental performance of a recycled and virgin polymer matrix in natural fiber composite production. The composites with the recycled polymer matrix exhibited significantly better environmental properties in non-automotive applications in the study. In automotive applications, the composites with the virgin polymer matrix had slightly better environmental performance due to their better functional performance compared to the composites with a recycled polymer matrix. Better functional performance led to decreased part weight, and therefore, to decreased fuel consumption during the automotive use phase. This advantage of the virgin polymer was entirely due to the differences in the functional performance of the polymers. If the recycled polymer had had equivalent performance with the virgin polymer, the environmental performance of the composite with the recycled polymer matrix would have been better also in the automotive applications. The environmental burden in the automotive applications is exceptionally heavily focused on the use phase, which can generate 70-90% of the total environmental impacts, unlike in non-automotive applications where the use phase rarely generates significant emissions.

Most of the current LCA studies concerning composites have been carried out after production. In the future, environmental assessment could be combined with micromechanical analysis done prior to the manufacturing phase, allowing optimization of the environmental performance already in the product design process. (Le Duigou & Baley 2014)

1.7 Economy of construction & demolition waste recycling

Economic incentives are seen as a major factor in the recycling of C&D waste. The economic viability of C&D waste recycling is determined by many factors, such as the costs of landfill deposition, transportation costs, recycling costs, and virgin raw material prices. Most of these factors vary depending on the location, and therefore, the economic viability must often be evaluated case by case. (Duran et al. 2006, Yuan et al. 2011)

The economic factors for a C&D waste producer and recycled material user often have to be determined separately, as their critical factors may differ from each other. From the C&D waste producers’ point of view, the main economic factors are landfill charges, transportation costs, labor costs, and recycling costs. Transportation costs, virgin raw material prices, and recycling costs are the main factors for a user of recycled materials. The recycling costs can be further divided to energy, equipment, and building costs. Usually building and equipment costs are considered as fixed costs, and energy costs are variable costs. (Duran et al. 2006, Kang & Schoenung 2006, Yuan et al. 2011)
Gate fee and product price are the main factors affecting the revenues of C&D waste recyclers. The producers of C&D waste pay a gate fee to the recycler when a given quantity of C&D waste is received at a recycling facility. Product price is the market price of the recycled products produced by the recycler. Both of these factors are strongly location-dependent. The gate fee of a recycling facility needs to be competitive with the gate fee of landfill deposition in order to create an incentive for the construction operator to bring the generated C&D waste into a recycling center instead of depositing it at a landfill. The product price is dependent on the supply and demand of materials, as well as the virgin raw material prices. The product price of recycled material should be considerably lower than the product price of virgin material in order to encourage the use of recycled materials. (Kartam et al. 2004, Zhao et al. 2010)

Naturally, economic viability is most likely to occur when the costs of taking C&D waste to a recycling facility are lower than the costs of landfill deposition, and when the price of the recycled material is lower than the price of virgin material. Economic viability is absolutely a critical factor in the formation of the markets for recycled materials. Currently, the lack of such markets is seen as a factor hindering the recycling rates of C&D waste. (Duran et al. 2006, Meinander et al. 2012)

The creation of the markets for secondary raw materials is important, as it would benefit both the society and the industry. A C&D producer could achieve lower waste disposal costs, and the recycled materials could offer savings in the raw material costs. The society could benefit from the lower environmental costs incurred. Therefore, it is clearly important to develop recycling systems where the supply and demand of secondary raw materials can be matched. (Duran et al. 2006, Meinander et al. 2012)
Aims of the study

2 Aims of the study

The main aim of this study was to study the impact of a recycled mineral filler on various properties of WPCs and to determine the critical factors affecting the formation of the properties. An estimation of the volume of mineral wool fiber waste generated in the European Union between years 2010-2020 was presented. Furthermore, the effect of fiber pre-treatment on the properties of WPCs were studied. Lastly, the environmental performance of a wood plastic composite containing recycled mineral fibers was assessed. The work is divided into categories as follows:

1) The volume of mineral wool fiber waste generated in the European Union (Paper I)
2) The effect of recycled mineral fibers on the functional properties of WPC
   2.1) Mechanical properties (Paper II)
   2.2) Moisture resistance properties (Paper II)
   2.3) Heat build-up and fire resistance properties (Paper III)
3) The effect of fiber pre-treatment on the functional properties of WPC (Paper IV)
4) The effect recycled mineral fiber utilization on the environmental properties of WPC (Paper V)

The synthesis presented here was based on these papers. The main idea was to give a comprehensive description of how the utilization of recycled mineral wool fibers as a filler in WPCs affects the functional and environmental properties of the composites. A flowchart of the structure of the thesis is presented in Figure 7.
Figure 7. Structure of the thesis.
3 Materials and methods

The materials and methods of the studies included in this thesis (Articles I-V) are presented in closer detail in each article. In this chapter the materials and methods are outlined only generally. General information of each article is presented below (Table 2).

<table>
<thead>
<tr>
<th>Paper</th>
<th>Materials</th>
<th>Methods</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>A review of current volumes of mineral wool waste in the European Union based on the Eurostat database</td>
<td>Literature review</td>
</tr>
<tr>
<td>II</td>
<td>Wood, PP, recycled mineral wool, MAPP</td>
<td>Determination of mechanical properties; flexural, tensile, impact, hardness, and moisture resistance; water absorption, thickness swelling and cyclic resistance</td>
</tr>
<tr>
<td>III</td>
<td>Wood, PP, recycled mineral wool, MAPP</td>
<td>Determination of heat-build up and fire resistance properties; heat release rate, total heat release and residual masses</td>
</tr>
<tr>
<td>IV</td>
<td>Wood, PP, recycled mineral wool, MAPP, γ-methacryloxypropyl trimethoxy silane</td>
<td>Determination of mechanical properties; flexural, tensile, impact, hardness, and moisture resistance; water absorption and thickness swelling</td>
</tr>
<tr>
<td>V</td>
<td>Collected and estimated data</td>
<td>Life cycle assessment (cradle-to-grave) of the composites</td>
</tr>
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The in-house manufactured research material consisted of 13 different WPCs. The compositions of the manufactured composites are shown in Table 3. Norway spruce (*Picea abies*) chips were used as the wood material in all composites. The wood chips used in the study were obtained from processing solid wood with crushers and hammermill apparatuses. The plastic matrix was PP in all composites. The mineral wool waste was rock wool waste from a rock wool manufacturing process, and it was obtained from the landfill of a rock wool plant. It was processed with crusher and hammer mill apparatuses before the composite manufacturing stage.

In the composite manufacturing stage, all the materials were agglomerated together prior to extruding with agglomeration apparatus consisting of a PLASMEC TRL100/FV/W
The bending test, flexural impact/Charpy and tensile properties were determined according to standards SFS-EN 310, SFS-EN ISO 179-1, and SFS-EN ISO 527-1, respectively. Brinell-hardness was measured according to standard SFS-EN 1534.

Moisture resistance under cyclic test conditions was determined according to standard SFS-EN 321, and three-point tests were carried out according to SFS-EN 310 standard after 3 weeks of soak/freeze/dry cycles. Water absorption and thickness swelling were determined according to standard SFS-EN 317.

The heat build-up in the WPCs was tested according to TS 15534 (Annex F). A total of three specimens per each type of composite were measured. A white infrared heat lamp having the nominal power of 250W (OsramSiccatherm FR 250W) was used. The distance between the lowest part of the downward-oriented lamp and the bottom of the box was 400 mm. The temperature of the composite measured at its bottom, and the increase of the temperature of the composite compared to ambient air (ΔTexp) were recorded with 20 second intervals by a digital thermometer equipped with a data logger. WPC material
Materials and methods

containing at least 25 g/kg furnace type carbon black, well dispersed and without any other pigmentation, was used as a control specimen.

A fire testing technology cone calorimeter was used for the reaction-to-fire tests according to ISO 5660. The samples were exposed in the horizontal orientation to an external heat flux of 50 kW/m² with the cone heater 25 mm above the sample surface. The samples were wrapped in aluminum foil covering the sides and the bottom. The exhaust system flow rate was 24 l/s. The cone data presented in this paper are the averages of three replicated experiments. During the tests, the following parameters were obtained: the heat release rate (HRR), total heat release (THR) and residual mass.

The size of the specimens for the bending tests and for moisture resistance under the cyclic test conditions was 450×50 mm. For the flexural impact tests, the size of the specimens was 80×10×5 mm. For measuring the tensile properties, the thickness of the dumbbell-shaped samples were 5 mm, and the width of the narrow part in the samples was 10 mm. The Brinell-hardness was tested from 75×75 mm samples. For the mechanical measurements, the average of 20 measurements was calculated, except for the tensile property (9–20 accepted measurements) and cyclic three-point tests (10 measurements). Samples with dimensions 75×75×5 mm were used for testing the heat build-up. The board specimens for the fire testing were 100mm × 100mm × 5mm in size.
4 Results and discussion

A brief description of the work and the findings of the individual articles are presented in the following. The sections are named according to the sense and purpose of the articles referred to in this work.

4.1 Mineral wool fibers in construction & demolition waste

Paper I presents the estimated volume of mineral wool waste generated in the European Union. Actual volumes of mineral wool waste from construction industry are complicated to estimate, as hardly any reliable data were available. However, Müller et al. (2009) have introduced a model to estimate the volume of mineral wool waste on the basis of mineral wool production volumes. The production volumes can be obtained from the Eurostat database (Eurostat 2015). In the model, the following assumptions were made: on average mineral wool has a service life of 30 years, and cutting scrap during new construction accounts for 5% of the products used. The estimation is based on a simple delay model, in which the distribution of lag time from construction to demolition was not considered, and a fixed lag time of 30 years is used. Müller et al. (2009) used the model to estimate mineral wool waste volumes in Germany; in this study the model was expanded to cover mineral wool waste volumes in the EU27 area.

Equation 1 was used in the model to estimate the amount of mineral wool waste generated annually:

\[ V_i = 0.95 \times P_{i-30} + 0.05 \times P_i, \]  

where \( V_i \) is the total volume of mineral wool waste generated in year \( i \), and \( P_i \) is the production volume of mineral wool (including both rock wool and glass wool) in year \( i \).

The first term of the equation is the volume of mineral wool waste generated by demolition activities, and the second term is the mineral wool waste generated by new construction. A 10-year forecast for 2010 and 2020 is presented in Figure 8.
Results and discussion

While the actual volume was a rather simple estimation, it was reasonable to assume that both mineral wool production and the volumes of generated waste grow annually. The accuracy of the model will improve over the years as more data are collected by the European Commission and published in the Eurostat database.

The total volume of C&D waste generated in the EU27 area in 2010 was reported to be 848.7 million tonnes (Eurostat 2015), and the estimated mineral wool waste volume in 2010 was 2.3 million tonnes. Accordingly, 0.3 % of all C&D waste generated was mineral wool.

4.2 The effect of recycled mineral fibers on the functional properties of wood polypropylene composites

4.2.1 Mechanical properties

In Paper II, the mechanical properties of wood plastic composites containing recycled mineral wool are presented. The investigated mechanical properties included flexural strength and modulus, tensile strength and modulus, impact strength, and Brinell hardness. The microstructure of the composites was examined with a scanning electron microscope (SEM). In this study, no fiber pre-treatment was applied, and MAPP was used as the coupling agent.
Results and discussion

Flexural strength

The flexural strength of the composites decreased when recycled mineral wool was added as the filler (Figure 9). The initial addition of 20% of recycled mineral wool decreased the flexural strength by 20% compared to the reference (MW0), where no mineral wool was used. Further increase of the recycled mineral wool content had less significant effects, as according to Duncan’s multiple range test the difference between composites MW20, MW30, and MW40 was not statistically significant at 95% confidence level.

![Figure 9. The initial flexural strengths of the studied composites. (II)](image)

Ashrafi et al. (2011) reported that the addition of E-glass fibers to WPCs reduced their flexural strength, as was the case with recycled mineral wool as well. Valente et al. (2011) reported a slight increase in flexural strength when wood flour was replaced with recycled glass fibers. Ashrafi et al. (2011) considered the fiber orientation and interfacial bonding of glass fibers and the polymer matrix to be the main factors affecting the flexural strength of composites. They noticed that glass fibers tended to orientate from normal to plunger motion. The same factors could be considered to affect the flexural strength of the mineral wool-filled composites, as in the SEM pictures it was observed that the fiber orientation in the manufactured composites was parallel with the direction of extrusion. Some fiber pull-outs, indicating non-optimal interfacial adhesion between the recycled mineral wool fibers and the polymer matrix, were also observed in the SEM pictures. The interfacial strength was also affected by the mechanical interlocking of the filler into the polymer matrix. Mineral wool fibers had smooth surfaces, which might lead to a lower degree of mechanical interlocking compared to wood fibers. Additionally, recycled mineral wool fibers had larger surface than wood chips, due to differences in particle size and shape. Therefore, the nonoptimal adhesion of the fillers to the matrix had a greater effect when a higher loading of recycled mineral wool fiber was used.
Flexural modulus

All the composites containing recycled mineral wool had lower flexural modulus than the reference composites containing no recycled mineral wool. The initial addition of 20% of recycled mineral wool decreased the flexural modulus by 14.9%. Increasing the recycled mineral wool content to 30% caused a further decrease of 5% in the flexural modulus. However, the composite with 40% of recycled mineral wool exhibited higher flexural modulus compared to the composites containing 20% and 30% recycled mineral wool (Figure 10). According to Duncan’s multiple range test there was a statistically significant difference between all the composites at 95% confidence level.

![Figure 10. The initial flexural moduli of the studied composites. (III)](image)

The flexural modulus of polymer composites may be affected by the strength of interfacial interactions between the fiber and the matrix, i.e., filler dispersion (Nuñez et al. 2003). Both the flexural and tensile moduli of the filler particles can also affect the flexural or tensile properties of the composites. Fillers with higher moduli can lead to higher moduli of the composites, provided that proper interfacial adhesion of the filler to the matrix is achieved (Premalal et al. 2002). The filler loading also affects the modulus of the composites, and usually an increase in the filler content leads to an increase in the modulus of the composite (Ashrafi et al. 2011).

In the studied composites, the total filler loading (wood chips + recycled mineral wool) was the same, 64% in all the composites. Therefore, the total filler loading did not affect the moduli of the composites. The differences in the moduli must have been caused by other factors: interfacial adhesion, filler dispersion, and the moduli of the wood and recycled mineral wool fibers. Mineral fibers typically have higher moduli compared to wood fibers. As no increase in the flexural moduli of the composites containing recycled mineral wool could be observed compared to the reference, as would be expected if there is no difference in the other factors, it must be concluded that the interfacial adhesion of recycled mineral wool fibers into the polymer matrix was weaker than the adhesion of wood fibers. The increasing effect of the higher modulus of the recycled mineral wool
fibers was at least partially lost due to the weak interfacial adhesion, though the composite containing 40 % of recycled mineral wool still had the highest modulus of the composites containing recycled mineral wool.

**Tensile strength**

The addition of recycled mineral wool into the composites decreased their tensile strength. The initial addition of 20 % of recycled mineral wool into the composite decreased the tensile strength by 21.4 % compared to the reference. The composite with 30 % of recycled mineral wool exhibited the lowest tensile strength value, 50.4 % lower than that of the reference. Further increase of the recycled mineral wool content to a total of 40 % caused 22.2 % increase in the tensile strength (Figure 11). Duncan’s multiple range test showed that there was a statistically significant difference between all the composites at 95 % confidence level.

![Figure 11. Tensile strengths of the studied composites. (II)](image-url)

Huuhiho et al. (2010b) showed that the addition of inorganic fillers to WPCs improved their tensile strength slightly. Rizvi and Semeralul (2008) also reported an increase in the tensile strength of WPCs when glass fiber was used as the filler. However, Gwon et al. (2010) found that increasing the mineral filler content in hybrid wood/mineral composites decreased the tensile strength of the composites. Similarly, Ashrafi et al. (2011) showed that an increase in the inorganic filler content had a negative effect on the tensile strength of composites.

The tensile strength of WPCs was affected by the same factors which influence the flexural strength: the strength of interfacial adhesion and fiber orientation. The tensile test results would also suggest that nonoptimal interfacial adhesion between the recycled mineral wool fibers and the polymer matrix has a severe impact on the strength properties of composites.
Tensile modulus

The tensile moduli of the composites followed the trend of tensile strength. The tensile modulus decreased by 8.8 % with the initial 20 % addition of recycled mineral wool and decreased further by 26.1 % when the recycled mineral wool content was increased to 30 %. When the total recycled mineral wool content reached 40 %, the tensile modulus improved by 33.1 % compared to the 30 % recycled mineral wool content. Figure 12 shows the tensile moduli of the studied composites. According to Duncan’s multiple range test there was no statistically significant difference in the tensile moduli between the composites containing 20 % and 40 % of recycled mineral wool.

![Figure 12. The tensile moduli of the studied composites. (II)](image)

Like flexural moduli, the tensile moduli of the composites were greatly affected by the amount of filler, as an increasing filler content increased the tensile modulus as well. In addition to the filler content, the tensile modulus could be affected by the modulus and size of the filler particles. Interfacial adhesion of the filler particles to the polymer matrix was not a major factor affecting the tensile modulus of the composites, but it has been shown that improved adhesion of the filler can also improve the tensile moduli of composites. (Fu et al. 2000, Ansari & Ismail 2009)

The total filler content remained constant in all the composites, and therefore, it should not have affected the modulus. Like in the flexural modulus, it was assumed that the differences in the moduli were caused by weaker adhesion of the mineral wool fibers to the polymer matrix.

Impact strength

The impact strength of the composites increased slightly when recycled mineral wool was added to the manufacturing process. The differences were rather small, as the addition of 20 % of recycled mineral wool to the composite increased the impact strength by 11.1 %.
The composite with 30% recycled mineral wool content had 6.3% higher impact strength than the reference, but 4.3% lower than the impact strength of the composite containing 20% of recycled mineral wool. The composite with 40% recycled mineral wool had the highest impact strength, 18.8% higher than the reference. Rather large standard deviations were observed in the measurement results, and the Duncan’s multiple range test showed statistically significant differences only between the reference and the composite containing 40% of recycled mineral wool. The impact strengths of the studied composites are shown in Figure 13.

![Figure 13. The impact strengths of the studied composites. (II)](image)

Cui and Tao (2009) found that the addition of glass fibers into WPCs can alter their impact strength. The impact strength was increased with the addition of L-glass fibers but decreased with S-glass fibers. The filler loading affects the impact strength of composites, usually an increasing filler loading leads to decreased impact strength. This is due to filler particles immobilizing the macromolecular polymer chains of the matrix, limiting their ability to deform freely and making the composite more ductile (Ansari & Ismail 2009). Furthermore, the interfacial adhesion of the filler to the polymer matrix and the particle size of the filler can affect the impact strength of composites (Premalal et al. 2002, Stark & Rowlands 2003).

Again, the total filler content of the studied composites stayed constant. As the composites containing recycled mineral wool had equivalent or higher impact strength compared to the reference, it can be partly attributed to the difference in the filler size: recycled mineral wool fibers have higher aspect ratio than wood chips, which could be a factor increasing the impact strength of the composites. As interfacial adhesion also contributes to the formation of impact strength properties, it can be assumed that part of the effect gained with the higher aspect ratio filler is could be lost due to the poorer interfacial adhesion.
Hardness properties

The surface of the composites became softer as recycled mineral wool was added to the manufacturing process. The composites with 20 %, 30 %, and 40 % of recycled mineral wool had 10.5 %, 16.5 % and 20.6 %, respectively, lower Brinell hardness than the reference (Figure 14). The differences in the Brinell hardness of the composites containing recycled mineral wool were rather small, as the Duncan’s multiple range test showed no statistically significant difference at 95 % confidence level between the composites containing 20 % and 30 % of recycled mineral wool or between the composites containing 30 % and 40 % of recycled mineral wool.

![Figure 14. The Brinell hardnesses of the studied composites. (II)](image)

The surface hardness value of composites can be used to predict their wear resistance, as hard materials have better wear and friction resistance (Georgopoulos et al. 2005). Badri et al. (2006) investigated the properties of natural fiber composites and found that increasing the filler load in the polymer matrix lowered the hardness of the composites. They concluded that the interfacial adhesion of the fibers had an effect on the surface hardness of the composites. It also has been shown that a high filler content can cause agglomeration of the filler particles, which has a negative effect on the surface hardness of composites (Deka & Maji 2010, Turku & Kärki 2014). Huuhilo et al. (2010b) noted that the Brinell hardness of mineral-filled WPCs was affected by the Moh’s hardness of the mineral filler. Harder fillers led to increased Brinell hardness of the composites.

In the studied composites, the decrease in the Brinell hardness of the composites when recycled mineral wool was incorporated into the manufacturing process could be explained by a weak interfacial adhesion of the fiber to the polymer matrix. While it could be assumed that the Moh’s hardness of recycled mineral wool fibers is higher than the Moh’s hardness of wood fibers, it seemed that the effect improving the Brinell hardness of the composites is overshadowed by the weak interfacial adhesion.
4.2.2 Moisture resistance properties

Cyclic resistance, thickness swelling and water absorption were tested to evaluate the moisture resistance properties of the composites.

Cyclic resistance

Cyclic testing is a material testing method where the material is exposed to repeated cycles of water immersion, freezing and thawing. In countries where outdoor temperatures are regularly below 0 °C, water immersion-freeze-thaw durability can provide valuable information to determine the expected service life of WPCs. (Butylina et al. 2011)

In this study, the impacts of cyclic testing on the flexural strength and modulus of the composites were determined. After the cyclic testing, the flexural strengths and moduli of the composites were lower than initially in all the cases. As shown in Figure 15, the flexural strength values of the composites were 76.7 %, 87.3 %, 75.8 %, and 78.1 % of the initial value for the composites containing 0, 20, 30, and 40 % of recycled mineral wool, respectively.

![Figure 15. Flexural strengths of the studied composites initially and after cyclic testing. (II)](image)

It can be seen in Figure 16 that the cyclic testing reduced the flexural moduli of the composites to 85.3 %, 86.4 %, 77.0 %, and 75.6 % of their initial values for the composites containing 0, 20, 30, and 40 % recycled mineral wool, respectively.
The moisture content (MC) of WPCs typically increases during cyclic testing. This increase in the moisture content is seen as the main factor affecting the strength properties of WPCs, and a greater moisture uptake results in greater loss of strength properties (Pilarski & Matuana 2006, Butylina et al. 2011). The weakening of the strength properties is due to the ability of moisture to change the structure and properties of the lignocellulosic fiber and the matrix, and the interface between them. When exposed to moisture, lignocellulosic fibers swell and develop shear stresses at the interface. Shear stress at the interface causes de-bonding of the fibers from the matrix, which again leads to reduction in the strength properties (Joseph et al. 2002, Butylina et al. 2011). Increase in the pore number and pore size may also be contributing factors in the weakening of mechanical properties during cyclic testing (Adhikary et al. 2010).

The addition of recycled mineral wool did not change the relative amount of weakening notably, except in the case of the composites containing 20% of recycled mineral wool, which had lower relative weakening than the other recycled mineral wool-containing composites. The weakening of the strength properties can be attributed to the increased moisture content of the composites and the de-bonding of the fibers from the matrix resulting from the increased MC.

**Thickness swelling and water absorption**

The thickness swelling of the composites decreased as the content of recycled mineral wool increased. The composites containing 40% of recycled mineral wool had 73.2% lower thickness swelling compared to the reference. The thickness swelling of the composites containing 20% and 30% of recycled mineral wool were 27.1% and 39.6% lower than the reference, respectively. It can be seen in Figure 17(a) that most of the composites had similar rates of swelling during the first 7 days of immersion, except for the composite containing 40% of recycled mineral wool, which had a clearly lower swelling rate. After 28 days there were significant differences between the thickness swellings of the composites.
The water absorption of the composites decreased with the addition of the recycled mineral wool filler. The water absorption of the composites containing 20%, 30%, and 40% of recycled mineral wool were 32.5%, 43.2%, and 67.4% lower than the water absorption of the reference composite, respectively. Figure 17(b) shows the water absorption of the composites.

Figure 17. Thickness swelling (a) and water absorption (b) of the studied composites. (II)

Butylina et al. (2011) identified water absorption as a major factor affecting the thickness swelling of mineral-filled WPCs. Adhikary et al. (2008) also recognized the porosity of
the matrix as an affecting factor in thickness swelling. Thickness swelling and water absorption seemed to follow a similar trend, the composites with the lowest water absorption also had the lowest thickness swelling. This would support the observation that the thickness swelling of WPCs is greatly affected by the water absorption.

Huuhilo et al. (2010a) studied mineral-filled WPCs and found that replacing part of the wood filler with a mineral filler lowered the water absorption of the composites. This could be explained by the hydrophilic nature of wood fibers and the hydrophobic nature of mineral fillers and the polymer matrix. Only the wood fraction of the composite is able to absorb water. Additionally, a mineral filler can reduce the water absorption by closing the pathways for water in the polymer matrix. Furthermore, if the wood fibers are completely encapsulated in the polymer matrix, water absorption is prevented, at least partially (Stark 2001). Mineral wool fibers are also hydrophobic, and therefore, their inclusion in the composite decreases the water absorption.

4.2.3 Heat build-up and fire properties

Paper III presents the heat build-up and fire properties of composites containing recycled mineral wool. The fire properties of WPCs are important because of the demands of fire protection in many applications (Li & He 2004). Applications in the construction industry or decking products are examples of WPC applications where the fire properties may be important. It has been shown that the fire properties of WPCs can be altered with mineral fillers (Nikolaeva & Kärki 2013). Temperature rise above the ambient air temperature (heat build-up) may have a significant effect on the dimensional stability of WPCs. Heat build-up is also a major factor with regard to comfort, for instance, when walking barefoot on deck boards.

Heat build-up

In heat build-up evaluation, the temperature of the specimen and the ambient temperature were recorded until the specimen reaches equilibrium temperature. The equilibrium temperature was considered to have been reached when the temperature of the test specimen did not change for five minutes. The heat build-up values were calculated from Equation 2:

\[
\Delta T = \frac{\Delta T_{\text{exp}(s)}}{\Delta T_{\text{exp}(c)}} \times \Delta T_r,
\]  

(2)

where \(\Delta T\) is the predicted heat build-up of the specimen due to heating by the sun, \(\Delta T_{\text{exp}(j)}\) is the temperature rise above the ambient temperature in the laboratory under the heat lamp, with \(j=s\) for the test specimen and \(j=c\) for the control specimen, and \(\Delta T_r\) is the
experimental heat build-up for the black control specimen under sun exposure, equaling 50 °C for horizontal position (e.g., decking) or 41 °C for vertical position (e.g., vertical siding).

In this study, the horizontal position was considered, giving ΔT, a value of 50 °C. The results of the heat build-up measurement are presented in Table 4.

Table 4. Results of heat-build up measurements (all temperatures in °C). (III)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>CB</th>
<th>MW0</th>
<th>MW20</th>
<th>MW30</th>
<th>MW40</th>
<th>MW50</th>
<th>MW64</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heat build-up ΔT</td>
<td>50.00</td>
<td>39.66</td>
<td>46.11</td>
<td>48.08</td>
<td>48.00</td>
<td>48.45</td>
<td>51.17</td>
</tr>
<tr>
<td>Temperature difference ΔTexp</td>
<td>58.2</td>
<td>46.2</td>
<td>53.7</td>
<td>56.0</td>
<td>55.9</td>
<td>56.4</td>
<td>59.6</td>
</tr>
<tr>
<td>Ambient temperature Ti</td>
<td>23.1</td>
<td>22.2</td>
<td>22.4</td>
<td>22.9</td>
<td>22.3</td>
<td>22.4</td>
<td>21.6</td>
</tr>
<tr>
<td>Sample thickness (mm)</td>
<td>4.8</td>
<td>4.9</td>
<td>4.7</td>
<td>4.9</td>
<td>4.6</td>
<td>4.8</td>
<td>4.8</td>
</tr>
<tr>
<td>Maximum temperature TM</td>
<td>81.3</td>
<td>68.4</td>
<td>76.1</td>
<td>78.9</td>
<td>78.1</td>
<td>78.8</td>
<td>81.1</td>
</tr>
</tbody>
</table>

The addition of recycled mineral wool increased the heat build-up of the composites. The initial addition of 20 % of recycled mineral increased the heat build-up by 16.3 % compared to the reference. Further increasing the recycled mineral wool content from 20 % to 30 % increased the heat build-up of the resulting composite by 4.3 %. Increasing the recycled mineral wool content above 30 % had a lesser effect on the heat build-up of the composites, as the differences between the composites containing 30-50 % of recycled mineral wool were less than 1 %. An increase of 5.6 % in the heat build-up of the composite was observed when the recycled mineral wool content increased from 50 % to 64 %. The composite containing 64 % of recycled mineral wool had the highest heat build-up value, 29.0 % higher than the reference, and 1.17 °C higher than the CB control sample.

The optical properties of composites have been identified as the key factor influencing the heat build-up (Martikka et al. 2012). CIELAB color coordinates are commonly used to determine the color of the WPCs. In the CIELAB model, three different color values: L*, a*, and b* are measured with a spectrometer (Fabiyi et al. 2008). The measured L*, a*, and b* color values for the studied composites are presented in Table 5.

Table 5. L*, a* and b* color coordinates of the studied composites. (III)

<table>
<thead>
<tr>
<th></th>
<th>CB</th>
<th>MW0</th>
<th>MW20</th>
<th>MW30</th>
<th>MW40</th>
<th>MW50</th>
<th>MW64</th>
</tr>
</thead>
<tbody>
<tr>
<td>L*</td>
<td>26.41</td>
<td>63.80</td>
<td>54.32</td>
<td>51.44</td>
<td>52.78</td>
<td>47.85</td>
<td>43.77</td>
</tr>
<tr>
<td>a*</td>
<td>0.03</td>
<td>4.48</td>
<td>0.91</td>
<td>0.52</td>
<td>-0.63</td>
<td>-0.53</td>
<td>-0.81</td>
</tr>
<tr>
<td>b*</td>
<td>0.03</td>
<td>26.19</td>
<td>19.88</td>
<td>18.43</td>
<td>15.91</td>
<td>13.91</td>
<td>11.65</td>
</tr>
</tbody>
</table>
Results and discussion

A regression analysis was performed to clarify the effect of each color coordinate on the heat build-up of the composites. The P value for the b* coordinate was above 0.1, and therefore it was not considered significant at a confidence level 90 % or higher. A simplified model for explaining the heat build-up of the composites could be expressed as Equation 3:

\[
\Delta T = 52.9613 - 1.57404 \times a^* - 0.096983 \times L^*.
\] (3)

The R\(^2\) value for the created model was 95.75, the standard error of estimation was 0.95 and the mean absolute error was 0.60. The heat build-up values predicted by the model and obtained from the measurements are presented in Figure 18.

![Figure 18. Predicted and measured heat build-up of the studied composites. (III)](image)

The obtained results are in agreement with the previous study by Martikka et al. (2012), though small differences can be found in the contribution of L* and a* values to the total heat build-up of the composites. This may be due to differences in absorption, reflection and transmittance of energy in the materials (Zubielewicz et al. 2011). Clark et al. (2007) reported a reflectance value of 0.5 for fiber glass-type mineral wool. Ljungdahl and Ribbing (1986) have measured the transmittance value of mineral wool between 2000 nm and 3000 nm wavelengths to be less than 0.01, and thus transmittance is not a major contributor in the heat build-up of recycled mineral wool-containing composites. Martikka et al. (2012) approximated that spruce wood has a reflectance value between 0.6 and 0.7, which is higher than the reflectance value of mineral wool. The lower reflectance value can contribute to the increased heat build-up of composites when part or all of the spruce wood is replaced with recycled mineral wool.
Fire performance

In the evaluation of fire performance, the heat release rates, total heat release and mass loss rates of the composites were studied. The heat release rates of the composites are presented in Figure 19. The sharp peak at the beginning of pyrolysis was characteristic for thinner samples, as the whole sample was pyrolysed at the same time (Schartel & Hull 2007). The second peak at the end of the pyrolysis was typical for wood materials, and an increased magnitude of the second peak could be observed in wood plastic composites with increasing wood content (Stark et al. 2010, Ayrilmis et al. 2012). The heat release rate is an important factor in determining the fire classification of materials with methods such as the single burning item (SBI) test (Messerschmidt & Van Hees 2000, Hakkarainen & Kokkala 2001).

![Figure 19. Heat release rates of the studied composites. (III)](image)

It can be seen in Figure 19 that the peak heat release rate did not change notably when recycled mineral wool was added to the composites. This would suggest that PP is the main contributor to the heat release rate of WPCs in the beginning of pyrolysis. The reference composite containing no recycled mineral wool had the highest peak at the end of pyrolysis. The peak at the end of pyrolysis decreased when recycled mineral wool was added to the composites. However, the decrease did not seem constant, as there were no significant difference in the intensity of the second peak between the composites containing 20 % and 30 % of recycled mineral wool. Similarly, the difference in the
Results and discussion

Intensity of the second peak between the composites containing 40% and 50% of recycled mineral wool was not notable. In the composite containing 64% of recycled mineral wool and no wood fibers, a secondary peak could not be observed.

The total heat release rates of the composites are presented in Figure 20. It can be seen that the addition of recycled mineral wool to the composites decreased their total heat release gradually. The composite containing 20% of recycled mineral wool had a 9.9% lower total heat release than the reference. After that every 10% addition of recycled mineral wool decreased the total heat release of the composite by 9.7-9.8% compared to the previous composite. The total heat release of the composite containing 64% recycled mineral wool was 19.1% lower than that of the reference.

![Figure 20. Total heat release of the studied composites. (III)](image)

The residual masses of the composites after the pyrolysis are presented in Table 6. It can be seen that the residual masses corresponded fairly well to the expected mass percentage of recycled mineral wool in the composites. This would suggest that recycled mineral wool was distributed rather evenly in the composites and did not burn in the pyrolysis process. The small differences in the mass percentages could be explained by the pyrolysis of organic substances, such as lubrication oils or glues.
Table 6. Original masses, residual masses and mass loss percentages of the studied composites.

<table>
<thead>
<tr>
<th></th>
<th>Original mass, g</th>
<th>Residual mass, g</th>
<th>Percentage of original mass</th>
</tr>
</thead>
<tbody>
<tr>
<td>MW0</td>
<td>50.3</td>
<td>0.4</td>
<td>0.80 %</td>
</tr>
<tr>
<td>MW20</td>
<td>54.5</td>
<td>10.6</td>
<td>19.45 %</td>
</tr>
<tr>
<td>MW30</td>
<td>55.0</td>
<td>13.6</td>
<td>24.73 %</td>
</tr>
<tr>
<td>MW40</td>
<td>58.6</td>
<td>21.5</td>
<td>36.69 %</td>
</tr>
<tr>
<td>MW50</td>
<td>60.9</td>
<td>27.6</td>
<td>45.32 %</td>
</tr>
<tr>
<td>MW64</td>
<td>72.3</td>
<td>44.1</td>
<td>61.00 %</td>
</tr>
</tbody>
</table>

Previous studies have shown that the effect of a mineral filler on the heat release rate of WPCs depends on the mineral used. Some minerals, such as talc, can reduce the peak values of the heat release rate, whereas minerals such as calcium carbonate may increase them. It is also possible that the mineral filler does not have a notable effect on the magnitude of the heat release rate peaks (Almeras et al. 2003, Nikolaeva & Kärki 2013). The effect of recycled mineral wool on the total heat release of the composites was similar to the effect of talc, which is a commonly used filler in WPCs. The residual masses of the studied composites were similar to composites containing other mineral fillers (Nikolaeva & Kärki 2013).

4.3 The effect of fiber pre-treatment on the properties of composites

In Paper IV, the effects of fiber pre-treatment with $\gamma$-methacryloxypropyl trimethoxy silane on the properties of the composites are reported. Different silane-based coupling agents are commonly used to modify the interfacial adhesion of both mineral and lignocellulosic fillers to the polymer matrix (Švab et al. 2005, Bouza et al. 2008). The silane-treated composites were compared to composites containing the same amount of recycled mineral wool but including MAPP as the coupling agent. Treatment solutions with three different silane concentration and composites with two different recycled mineral wool contents, 20 % and 40 %, were studied. As the fundamental mechanics affecting the properties of WPCs were already discussed in detail in Chapter 4.2, the focus of this chapter is on how the silane-treated composites differ from the composites containing MAPP as the coupling agent.

4.3.1 Mechanical properties

The investigated mechanical properties included: flexural strength and modulus, tensile strength and modulus, impact strength, and Brinell hardness.

Flexural properties

The silane-treated composites presented generally lower flexural strength values than the composites with MAPP as the coupling agent (Figure 21). The composite with 3 % silane...
Results and discussion

treatment and 20 % of recycled mineral wool content had clearly the highest flexural strength of the silane-treated composites, which was comparable with the composite containing 20 % of recycled mineral wool and MAPP, as Duncan’s multiple range test did not show a statistically significant difference between the two at 95 % confidence level. The rest of the silane-treated composites had clearly weaker flexural strengths compared to their MAPP-containing counterparts.

Figure 21. Flexural strengths of silane-treated (MW20 & MW40) and MAPP-containing (MW20M3 & MW40M3) composites. (IV)

The strength of interfacial adhesion and fiber orientation, as well as the size and morphology of the filler particles are the main factors affecting the flexural strength of WPCs. What has been said earlier concerning the properties of MAPP-containing composites on the effects of these factors on the properties of composites could also be applied here. Furthermore, the scanning electron microscopy (SEM) analysis of the silane-treated composites revealed that the surfaces of the recycled mineral wool fibers did not have any polymer adhered to them, indicating that the interface between the fibers and the polymer matrix had been the weakest point in the composites, as the fracture path had progressed through the interface. The SEM pictures are shown in Figure 22.

Possible reasons for the weak interfacial adhesion of the fillers when using the silane coupling agent were nonoptimal grafting of silanol monomers or oligomers to the filler surfaces, lack of treatment solution absorption into the cell walls of the wood fibers, and excessive polymerization of silanol monomers during the treatment process.
Figure 22. Fracture surfaces of the composites: (a) MW20S1, (b) MW20S3, (c) MW20S5, (d) MW40S1, (e) MW40S3, (f) MW40S5, (g) MW20M3, and (h) MW40M3. (IV)
Results and discussion

Flexural modulus

The flexural moduli of the silane-treated composite containing 20 % of recycled mineral wool were higher than the flexural modulus of the composite with the corresponding amount of mineral wool and MAPP as the coupling agent. However, in the case of the composite containing 40 % of recycled mineral wool, the flexural moduli of the silane-treated composite were the same (1 % silane concentration) or lower than the flexural modulus of the composite with MAPP. The flexural moduli of the composites are shown in Figure 23.

![Figure 23. Flexural moduli of silane-treated (MW20 & MW40) and MAPP-containing (MW20M3 & MW40M3) composites. (IV)](image)

The flexural modulus of WPCs was affected by the strength of interfacial interactions between the fiber and the matrix, filler dispersion, filler loading, and the mechanical properties of the filler fiber. In the composites containing MAPP as the coupling agent, the higher modulus of the composite containing 40 % of recycled mineral wool could be explained by the higher modulus of the recycled mineral wool compared to wood fibers. In silane-treated composites the decrease in the flexural modulus with increasing recycled mineral wool content could be attributed to a less optimal adhesion of the recycled mineral wool fibers to the matrix compared to the MAPP-containing composites. The silane-treated composites containing 20 % of recycled mineral wool had higher modulus compared to their counterpart containing MAPP. This could be attributed to the slightly higher total filler content of the silane-treated composites compared to the composite containing MAPP and the better interfacial adhesion of the wood fibers to the matrix compared to recycled mineral wool fibers. In the silane-treated composites containing 40
% of recycled mineral wool, the advantage gained from the higher total filler content was lost due to the less optimal interfacial adhesion of the recycled mineral wool fibers.

**Tensile strength**

The tensile strengths of the silane-treated composites followed a similar trend as observed in the flexural strengths of the composites. The composites containing 20 % of recycled mineral wool had higher tensile strengths than the composites containing 40 % of recycled mineral wool. Of the composites containing 20 % of recycled mineral wool, the composite with MAPP as the coupling agent and the composite treated with 3 % silane concentration had the highest tensile strengths. According to Duncan’s multiple range test, there was no statistically significant difference between the two at 95 % confidence level. The differences in the tensile strengths of the composites containing 40 % of recycled mineral wool were small, as the only statistically significant difference could be observed between the composite containing MAPP and the composites treated with 3 % silane concentration. The tensile strengths are presented in Figure 24.

![Tensile strengths of silane-treated (MW20 & MW40) and MAPP-containing (MW20M3 & MW40M3) composites. (IV)](image)

The tensile strength of composites was affected largely by the same factors which affected the flexural strength. The nonoptimal adhesion of recycled mineral wool fibers, together with their relatively large surface area had a negative effect on the tensile strengths of the composites containing 40 % of recycled mineral wool. The composites containing the silane coupling agent had lower tensile strengths compared to the composites with the same recycled mineral wool content and MAPP as the coupling agent. This was a further
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indication of the fact that MAPP provides better adhesion of fillers to the polymer matrix compared to the silane coupling agent.

**Tensile modulus**

The filler content was the main factor affecting the tensile modulus of WPCs. The tensile modulus was also affected by the quality of interfacial adhesion between the filler and the matrix, but the effect was minimal. The silane-treated composites containing 20 % of recycled mineral wool had a significantly higher tensile modulus compared to the MAPP-containing composite. Figure 25 shows the tensile moduli of the composites. In the composites containing 40 % of recycled mineral wool, only the composite treated with 3 % silane concentration had a significantly higher tensile modulus than the other composites.

The higher total filler content was the main explaining reason for the higher tensile moduli of the silane-treated composite containing 20 % of recycled mineral wool. Again, the nonoptimal interfacial adhesion of the recycled mineral wool fibers in the composite containing 40 % of recycled mineral wool hindered the tensile moduli of these composite. The weak interfacial adhesion was also the likely reason why the silane treated composites containing 20 % of recycled mineral wool had higher tensile moduli than the silane-treated composites containing 40 % of recycled mineral wool, despite the recycled mineral wool fibers having higher moduli compared to wood fibers.
Impact strength

The silane-treated composites containing 20% of recycled mineral wool had a higher impact strength than the MAPP-containing composites with the same volume of recycled mineral wool. The impact strength values for the composites containing 40% of recycled mineral wool were very close to each other, regardless of the coupling agent used. The impact strengths of the composites are shown in Figure 26. Overall, the differences in the impact strengths of the silane-treated composites were small, as only the composite with 20% of recycled mineral wool and 3% silane treatment was significantly different from the others. None of the composites containing 40% of recycled mineral were significantly different from each other.

![Figure 26. Impact strengths of silane-treated (MW20 & MW40) and MAPP-containing (MW20M3 & MW40M3) composites. (IV)](image)

The strength of interfacial adhesion between the filler particles and the polymer matrix, filler particle loading and particle size were the main factors affecting the impact strength of composites. Interfacial adhesion was probably the main reason why the composite containing 20% of recycled mineral wool and treated with 3% silane concentration had better impact strength than the other composites. The composites containing 20% of recycled mineral wool and MAPP as the coupling agent contained wood fibers with slightly shorter fiber length than the other composites, which could explain the lower impact strength compared to the other composites. It has been shown that in some cases the impact strength of composites can increase when the filler size increases (Fu et al. 2008).
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**Hardness properties**

The silane-treated composites had lower surface hardness values than their MAPP-containing counterparts. The composites containing 40% of recycled mineral wool had lower surface hardness than the composites containing 20% of recycled mineral wool. The Brinell hardnesses of the composites are presented in Figure 27.

![Figure 27. Brinell hardness of silane-treated (MW20 & MW40) and MAPP-containing (MW20M3 & MW40M3) composites.](image)

The main factors affecting the surface hardness of composites were the interfacial adhesion of the filler particles, the properties of the filler particle and the total filler loading. The higher surface hardness of the MAPP-containing composites could be explained by the slightly lower total filler content and better adhesion of the filler particles to the polymer matrix. It was observed that increasing the mineral filler loading in the composites led to a decrease in the Brinell hardness values of the composites for both types of coupling agents. This could be attributed to the weaker adhesion of recycled mineral wool compared to wood fibers.

**4.3.2 Water absorption and thickness swelling properties**

The water absorption of the silane-treated composites was slightly higher compared to the composites with the same recycled mineral wool content and MAPP as the coupling agent. The composites containing 40% of recycled mineral wool absorbed less water than the composites containing 20% of recycled mineral wool. The silane treatment percentage did not affect the water absorption of the composites notably, although in the
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In the case of the composites with 40% of recycled mineral wool, the composites treated with 3% silane concentration absorbed more water than the others. Figure 28(a) shows the water absorption rates of the composites.

Water absorption in wood fibers is mainly due to hydrogen bonding of the water molecules into hydroxyl groups on the cell walls of the wood fibers. Silane treatment can reduce the water absorption of wood fibers, as the silane coupling agents react with the same hydroxyl groups of the wood fibers, leaving less active groups to form hydrogen bonds with the water molecules (Ichazo et al. 2001). Replacing the hydrophilic wood with hydrophobic recycled mineral wool led to lower water absorption. The silane-treated composites had higher water absorption as they contained slightly more wood fiber due to the difference in the way the silane coupling agent and MAPP were applied to the composites. The percentage of the silane treatment did not have a major effect on the water absorption of the composites.

![Graph](image)

Figure 28. The (a) water absorption and (b) thickness swelling of silane-treated (MW20S1-5 & MW40S1-5) and MAPP-containing (MW20M3 & MW40M3) composites. (IV)
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The composites containing 20 % of recycled mineral wool swelled more than the composites containing 40 % of recycled mineral wool. In the case of the silane-treated composites with 20 % of recycled mineral wool, the composites treated with 1 % and 5 % silane concentration swelled more than the composites treated with 3 % concentration. The composite containing 20 % of recycled mineral wool and MAPP as the coupling agent swelled more than the 3 % silane-treated composites but less than the 1 % and 5 % silane-treated composites. The silane-treated composites containing 40 % of recycled mineral wool swelled considerably more than the MAPP-containing composite with the same amount of recycled mineral wool. The concentration of the silane treatment did not have a major effect on the thickness swelling of the composites with a 40 % recycled mineral wool content. The thickness swelling values of the composites are presented in Figure 28(b).

The thickness swelling of the composites can be related to both water absorption and the mechanical properties of the composites. All the silane-treated composites with 20 % of recycled mineral wool had almost the same water absorption, but the thickness swelling of the composite treated with 3 % silane concentration was notably lower than the swelling of the composites treated with 1 % or 5 % concentration. It also had clearly superior mechanical properties compared to the other silane-treated composites. However, the composites with 20 % of recycled mineral treated with 3 % silane concentration had higher water absorption than the composite with the same amount of recycled mineral wool and MAPP as the coupling agent, but still lower thickness swelling despite there being only small or no significant differences in the mechanical properties. For the composites containing 40 % of recycled mineral wool, the composites with higher water absorption had also higher thickness swelling, but the differences in the mechanical properties were also smaller than the differences between the silane-treated composites containing 20 % of recycled mineral wool.

4.4 Environmental impacts of the utilization of recycled mineral fiber

In Paper V the environmental impacts of the use of recycled mineral wool as a filler in wood plastic composites were assessed. The obtained results were compared to glass fiber, which is a traditional mineral filler used in wood plastic composites. A 20 % mineral filler content was used in both scenarios. The environmental profile of a wood plastic composite with no mineral filler was also presented as a reference. The components of the composites in each scenario, as well as abbreviations of the scenario names are presented in Table 7.

LCA was conducted according to the requirements and guidelines presented in standards SFS-EN ISO 14040 and SFS-EN ISO 14044. The assessment covered the whole life cycle of the products, from raw material production to end-of-life disposal (cradle-to-grave approach). Two different end-of-life options for composite waste were also evaluated: incineration for energy and landfill disposal.
Table 7. Components of the studied composites as weight percentages, and abbreviations of the scenario names. (V)

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Wood flour</th>
<th>Glass fiber</th>
<th>Recycled mineral wool fiber</th>
<th>Virgin PP</th>
<th>Recycled PP</th>
</tr>
</thead>
<tbody>
<tr>
<td>30VPP</td>
<td>70</td>
<td>0</td>
<td>0</td>
<td>30</td>
<td>0</td>
</tr>
<tr>
<td>20GF</td>
<td>50</td>
<td>20</td>
<td>0</td>
<td>30</td>
<td>0</td>
</tr>
<tr>
<td>20MW</td>
<td>50</td>
<td>0</td>
<td>20</td>
<td>30</td>
<td>0</td>
</tr>
</tbody>
</table>

PP refers to polypropylene

The classification and characterization of the results was done according to the CML 2001 method (Guinée 2001). Weighting or normalization was not used in the study. The assessed impact categories were global warming potential during a 100-year time period (GWP), acidification potential (AP), eutrophication potential (EP), abiotic depletion of fossil resources (ADP (fossil)), and abiotic depletion of elemental resources (ADP (elemental)).

4.4.1 Global warming potential

The global warming potential in the different scenarios is shown in Figure 29. Scenario 20GF had the highest GWP in both end-of-life options. The global warming potential of 20MW was slightly lower than that of 30VPP, mainly because less emissions were generated in the end-of-life actions. In scenario 20MW the composite contained a greater amount of inert material compared to the composites in the 30VPP scenario, and therefore, the emissions from the incineration of the composite waste were reduced.

The end-of-life options had a great effect on the GWP of the composites. Incineration generated more GWP-related emissions than landfill deposition of composite waste. The landfill deposition of composites generated about 50 % less emissions than the incineration of composite waste in all scenarios.
In this study, 96% of the GWP-related emissions from landfill deposition could be attributed to degradation of the wooden fraction of the composite, as the plastic fraction of the composite degrades very slowly. According to Finnvelden et al. (1995) 1-5% of the plastic fraction degrades in a 100-year period. The inert fraction generated less than 1% of the GWP-related emissions in landfill deposition. In the incineration process, the polymer fraction was degraded and its share of the GWP-related emissions varied between 45-53%, depending on the scenario. In all scenarios, the plastic and wooden fraction together were responsible for 98% of the GWP-related emissions generated during the incineration of composite waste, including transportation of the waste to the incineration facility.

Witik et al. (2013) have studied the environmental impacts of different end-of-life options for polymer composites and obtained similar results regarding the GWP of polymer composites. Lazarevic et al. (2010) have analyzed a number of studies concerning the recycling options of plastic waste and note that when landfill deposition and incineration of plastic waste were compared, the GWP of landfill deposition was lower in most studies. They also note that if the examination period is longer than 100 years, the GWP advantage of landfill deposition can be reduced, as more of the plastic fraction will degrade. Carpenter et al. (2013) have studied the end-of-life options for wooden C&D debris and found that the incineration of wooden debris had a positive GWP compared to landfill deposition.

4.4.2 Acidification potential

The VPP scenario with incineration of the composite achieved the lowest acidification potential, followed by the 20MW scenario, also with incineration of the composite waste.
Scenario 20GF had the highest AP in both end-of-life options. The production of energy needed to operate the composite production line was a major source of AP-related emissions, its share varied between 21.7-54.6 % of the total AP, depending on the scenario and the end-of-life option. Scenario 20GF was burdened by a high AP of glass fiber production, as well as a slight disadvantage from lower energy production from composite waste incineration compared to 30VPP. A similar disadvantage existed between scenarios 20MW and 30VPP, where scenario 20MW required production of energy to replace the slightly lower energy acquisition from composite waste incineration. The acidification potential of the scenarios is presented in Figure 30.

The difference between zero-burden recycled fuels and replacing energy production technologies can be most clearly seen in the comparison of the end-of-life options, where incineration was clearly a favorable option in all the scenarios presented. The wooden fraction was a major contributor of AP-related emission in both end-of-life options, its share was 27-36 % in incineration and 62-73 % in landfill deposition, depending on the scenario. The plastic fraction had a contribution of 18-19 % in incineration and 18-21 % in landfill deposition. The inert fraction caused 7 % of AP-related emissions in incineration and 6 % in landfill deposition. The transportation of the waste had a notable effect on the AP in the end-of-life options. In landfill disposal, where the waste transportation distance was shorter (40 km), the share of transportation was 9-12 % of the total AP. In incineration, where the transportation distance was 250 km, the share of AP-related emission originating from transportation was 46-48 %.

Figure 30. Acidification potential of the different scenarios and end-of-life options. (V)
Corbière-Nicollier et al. (2001) have compared the environmental properties of polymer composites filled with glass fibers and CR fibers. It was shown in their study that the composites containing CR had a lower AP compared to the glass fiber-filled composite. Similarly, in the present study the wood fiber had a lower AP compared to glass fibers. Lazarevic et al. (2010) have reviewed studies concerning incineration and landfill deposition of plastic waste and found that in most cases the AP of incineration was lower than the AP of landfill deposition. It should be kept in mind that in the present study, only 18-21 % of AP-related emissions originated from the plastic fraction of the composites, therefore the total results cannot be directly compared to the results obtained for a pure plastic fraction. However, no studies discussing the AP of the end-of-life options for wood waste could be found.

### 4.4.3 Eutrophication potential

Scenario 30VPP with incineration as the end-of-life option had the lowest EP, followed by scenario 20MW, again with incineration as the end-of-life option. Scenario 20GF had the highest EP. The eutrophication potential of the different scenarios is presented in Figure 31.

![Figure 31. Eutrophication potential of the different scenarios and end-of-life options. (V)](image)

Energy production was a major source of EP-related emissions, which largely explained the good EP of the 30VPP scenario, as no replacement energy was needed in the scenario. Production and transportation of both virgin PP and glass fibers generated a relatively large amount of EP-related emissions. The share of virgin PP of the total EP varied between 9-24 %, depending on the scenario. Glass fiber accounted for 24-38 % of the
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Total EP, depending on the end-of-life option. Recovery and transportation of mineral wool fibers generated 3-6% of the total EP-related emissions.

When incineration was chosen as the end-of-life option, scenario 30VPP had lower EP compared to 20MW, because no replacement energy was needed. When the end-of-life option was landfill deposition, scenario 20MW performed better due to the lower wood material content of the composite. Compared to recycled mineral wool, the wooden material generated more EP-related emissions when deposited to a landfill.

The landfill deposition of composite waste generated considerably more EP-related emissions than incineration. Furthermore, more replacement energy was needed in the landfill deposition option. In incineration, transportation of the composite waste to the incineration plant generated 57% of the total EP-related emissions. Incineration of the wooden material fraction generated 21-29% of the EP-related emissions. 15-16% of the EP-related emissions during the incineration of composite waste originated from the plastic material fraction. Incineration of inert waste generated 7% of the EP-related emissions in the scenarios where recycled mineral wool or glass fiber was used. In landfill deposition, only 5% of the EP-related emissions were caused by the transportation of the composite waste. The wooden material fraction was the biggest source of EP-related emissions in landfill deposition, accounting for 56-65% of the total EP-related emissions. The plastic material fraction was the second biggest source, responsible for 31-37% of the total EP-related emissions in landfill deposition. Inert waste caused 2% of the total EP-related emissions in landfill deposition in the scenarios where either recycled mineral wool or glass fiber were used.

Luz et al. (2010) have studied the end-of-life options for sugarcane bagasse-PP composites and found that the EP of composite waste incineration was lower than the EP of landfill deposition. Lazarevic et al. (2010) state that most of the studies concerning recycling options for plastic waste report incineration to have a lower EP compared to landfill deposition.

4.4.4 Abiotic depletion

ADP of fossil resources was mainly caused by the production of virgin PP and energy. Transportation, harvesting of resources and waste sorting also caused ADP (fossil), but their role was less significant. The production of virgin PP caused 54-69% of the total ADP (fossil). Scenarios 20MW and VPP had almost the same performance as far as ADP (fossil) is concerned. The production phase of scenario VPP had a slightly higher ADP (fossil), but the difference was evened out in the end-of-life phase, where scenario 20MW required production of replacement energy. Scenario 20GF was again burdened by the production and transportation of glass fibers, which accounted for 14-15% of the total ADP (fossil) in the scenario. The recovery and treatment of recycled mineral wool in scenario 20MW caused less than 1% of the total ADP (fossil) of the scenario. The ADP (fossil) of the scenarios is presented in Figure 32.
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The ADP (fossil) caused by the incineration process of the composite waste was roughly the same as the ADP (fossil) caused by landfill deposition, and in addition, replacement energy was needed in the landfill deposition option. This explains the difference in the total ADP (fossil) between the end-of-life options. The ADP (fossil) caused by the replacement energy production was around 12% of the total ADP (fossil) in all the scenarios.

Comparisons with other studies are complicated because the abiotic depletion of fossil and element resources are rarely separated. The review of studies concerning the recycling of plastic waste by Lazarevic et al. (2010) reports that incineration causes less ADP compared to landfill deposition. Luz et al. (2010) report that the ADP of landfill deposition is lower compared to the incineration of sugarcane bagasse-PP composites. The difference in the results can be simply due to different process parameters, such as transportation distances or the energy production technologies chosen.

The production of glass fibers was by far the greatest cause of abiotic depletion of element resources, it was responsible for 99% of the ADP (element) in scenario 20GF (Figure 33). Incineration as the end-of-life option had a slight advantage compared to landfill deposition, as it is possible to receive some metals from the bottom ash generated in the incineration.

Figure 32. Abiotic depletion of fossil resources of the different scenarios and end-of-life options. (V)
Figure 33. Abiotic depletion of elements of the different scenarios and end-of-life options. (V)
4.5 Synthesis of the study

The impacts of the volume of recycled mineral wool filler and different treatments on the functional properties of the composites are presented in Table 8. Improvement in a property is marked with the + sign while decline in a property is marked with the - sign. The number of signs represents the magnitude of the effect. In the case of heat build-up, increase in the heat build-up was considered as a negative effect, but there might be applications where it could be considered as a positive effect. The silane-treated composites were compared to the case where a similar volume of recycled mineral wool and maleated polypropylene as the coupling agent were used.

Table 8. Impact of the volume of recycled mineral wool filler and different treatments on composites. N/A=not available.

<table>
<thead>
<tr>
<th>Composite/treatment</th>
<th>Mechanical properties</th>
<th>Moisture resistance</th>
<th>Heat build-up</th>
<th>Fire resistance properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>20 % of recycled mineral wool</td>
<td>-</td>
<td>+</td>
<td>-</td>
<td>+</td>
</tr>
<tr>
<td>30 % of recycled mineral wool</td>
<td>- -</td>
<td>++</td>
<td>- -</td>
<td>++</td>
</tr>
<tr>
<td>40 % of recycled mineral wool</td>
<td>-</td>
<td>+++</td>
<td>- -</td>
<td>++</td>
</tr>
<tr>
<td>50 % of recycled mineral wool</td>
<td>N/A</td>
<td>N/A</td>
<td>- -</td>
<td>++</td>
</tr>
<tr>
<td>64 % of recycled mineral wool</td>
<td>N/A</td>
<td>N/A</td>
<td>- - -</td>
<td>+++</td>
</tr>
<tr>
<td>20 % of recycled mineral wool, 1 % silane treatment</td>
<td>-</td>
<td>- -</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>20 % of recycled mineral wool, 3 % silane treatment</td>
<td>+</td>
<td>-</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>20 % of recycled mineral wool, 5 % silane treatment</td>
<td>-</td>
<td>- -</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>40 % of recycled mineral wool, 1 % silane treatment</td>
<td>-</td>
<td>- -</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>40 % of recycled mineral wool, 3 % silane treatment</td>
<td>- -</td>
<td>- -</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>40 % of recycled mineral wool, 5 % silane treatment</td>
<td>- -</td>
<td>- -</td>
<td>N/A</td>
<td>N/A</td>
</tr>
</tbody>
</table>

The addition of recycled mineral wool into the composites had a negative effect on the mechanical properties of the wood plastic composites. When MAPP was used as the coupling agent, composites with 30 % of recycled mineral wool performed worse than the composites containing 20 % or 40 % of recycled mineral wool. There was no notable difference in the overall mechanical performance of the composites containing 20 % or 40 % of recycled mineral wool. The composite containing 20 % of recycled mineral wool performed slightly better in the case of strength properties but had worse performance in flexural and tensile moduli. Ashrafi et al. (2011), among others, have obtained rather similar results when studying the addition of glass fibers into WPCs.
Silane treatment, compared to MAPP as the coupling agent, did not generally provide better mechanical properties. Only in the case where 20% of recycled mineral wool and a 3% concentration of silane treatment solution was used did the overall mechanical properties improve. The increase was due to improvement in both flexural and tensile moduli, as well as impact strength. The flexural and tensile strengths were comparable to the situation where MAPP was used, and the Brinell hardness exhibited a decrease. Therefore, the effect on the overall performance could be seen as positive. Santos et al. (2009) compared the effect of vinyltriethoxysilane and MAPP on the elastic (tensile) modulus of coir-filled PP composites and found that vinyltriethoxysilane provided a higher modulus when the coupling agent concentration was higher than 0.5 wt-% of the mass of the polymer matrix. Similar results were obtained in the present study where the silane treatment in certain cases improved the flexural and tensile moduli of the composites. Das et al. (2010) compared a silane coupling agent and MAPP in fly-ash filled PP composites. They conclude that the silane-based coupling agent provided slightly better flexural properties than MAPP, due to good reactivity between fly ash and the silane coupling agent. It was also shown in their study that the coupling agent concentration had a major effect on the properties of the composites. When comparing these results to the results of the present study, the differences could be due to the different chemical reactivity of the filler particles and different coupling agent concentrations used.

The moisture resistance properties of the composites were significantly improved when recycled mineral wool was added into the composites. Clear improvements were observed in the composites containing 20% of recycled mineral wool, and the magnitude of the improvements grew as the volume of recycled mineral wool in the composites increased. Improvements were observed in both water absorption and thickness swelling of the composites. The cyclic resistance of the composites did not change notably, except in the case of the composite containing 20% of recycled mineral wool, which had better cyclic resistance compared to the other composites. The results obtained concerning the impact of recycled mineral wool fibers on the moisture resistance properties were well in line with the results found in the literature. For example, Shakert and Ghasemian (2010) obtained corresponding results with recycled newspaper/glass fiber/PP composites, as did Panthapulakkal and Sain (2007) with hemp/glass fiber/PP composites.

The silane-treated composites had worse moisture resistance than the composites with the same mineral wool content but MAPP as the coupling agent. The composite containing 20% of recycled mineral wool treated with a 3% silane concentration exhibited the best moisture resistance properties of all the silane-treated composites, mainly because of advantages in the thickness swelling of the composite, but even then the composites containing MAPP as the coupling agent had superior moisture resistance performance. Previous studies comparing the effects of a silane coupling agent and maleated polymer in regard to moisture resistance properties are scarce, but Larivière et al. (2005) have shown that a silane-based coupling agent caused rather mild improvement in the moisture resistance properties of glass fiber/PP composites, and Kushwaha and Kumar (2010) observed similarly slight improvement with silane-based coupling agents in NaOH-treated bamboo/epoxy composites, whereas Mohebby et al. (2010) found that MAPP
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improved the moisture resistance properties of a wood flour/glass fiber/PP composite significantly.

The heat build-up of the composites increased, which here was considered a negative effect. The heat build-up increased along with the increasing volume of recycled mineral wool in the composites, though the changes between 30 % and 50 % recycled mineral wool content were less severe. The results obtained in the present study showed only minor differences compared to the results obtained by Martikka et al. (2012).

The fire resistance properties of the composites also improved when recycled mineral wool was added into the composites. As the addition of recycled mineral wool did not affect the peak heat release rate of the composites notably, the total heat release rose as the dictating factor in the fire resistance properties. A significant improvement in the total heat release was observed with the initial addition of 20 % of recycled mineral wool. A smaller improvement was observed when the volume of recycled mineral wool was increased to 30 %. When the recycled mineral wool content rose from 30 % to 50 %, the improvements in fire performance were rather small. Raising the recycled mineral wool content from 50 % to 64 % again produced a relatively great improvement in the fire resistance properties. Giancaspro et al. (2008) studied the heat release rates of a sawdust/carbon & glass fiber/inorganic matrix and noticed that the addition of the carbon & glass fiber mixture did not lower the peak heat release rates of the composites. Previous results obtained by Nikolaeva and Kärki (2013) showed that many inorganic fillers in wood plastic composites had a similar effect as recycled mineral wool fibers on the peak heat release rate and total heat release of wood plastic composites.

Table 9 shows the impacts of mineral fillers on the environmental performance of the composites compared to wood fiber as the filler. Recycled mineral wool had superior performance compared to glass fibers in every impact category assessed. The results obtained for glass fiber compared to wood fibers were in agreement with the previous studies by Corbière-Nicollier et al. (2001) and Joshi et al. (2004). No previous studies concerning the LCA of recycling of inorganic filler materials in wood plastic composites were available, but Rajendran et al. (2012) have shown that utilizing recycled raw materials in wood plastic composites can have environmental benefits.

<table>
<thead>
<tr>
<th>Table 9. Impact of the mineral filler materials on the environmental impacts of composites. (V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Filler material</td>
</tr>
<tr>
<td>---------------------</td>
</tr>
<tr>
<td>Recycled mineral wool</td>
</tr>
<tr>
<td>Glass fiber</td>
</tr>
</tbody>
</table>

GWP refers to global warming potential, AP refers to acidification potential, EP refers to eutrophication potential, ADP (fossil) refers to abiotic depletion of fossil resources, and ADP (elements) refers to abiotic depletion of elemental resources.
In Table 10, the environmental performance of the end-of-life options are presented. As only two different end-of-life options were studied, the option with better environmental performance is marked with the + sign and the option with worse environmental performance is marked with the - sign. The environmental performance is reported separately for each studied impact category. Incineration of composite waste was a favorable end-of-life option in all the impact categories except for global warming potential, where landfill deposition had better performance.

Table 10. Environmental performance of the end-of-life options. (V)

<table>
<thead>
<tr>
<th>End-of-life option</th>
<th>GWP</th>
<th>AP</th>
<th>EP</th>
<th>ADP (fossil)</th>
<th>ADP (elements)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Incineration</td>
<td>-</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>Landfill deposition</td>
<td>+</td>
<td>-</td>
<td>-</td>
<td>-</td>
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GWP refers to global warming potential, AP refers to acidification potential, EP refers to eutrophication potential, ADP (fossil) refers to abiotic depletion of fossil resources, and ADP (elements) refers to abiotic depletion of elemental resources.

Comparisons to other studies regarding the end-of-life options can be problematic due to the fundamental effect of the parameters chosen in the studies. Furthermore, relatively few previous studies have assessed the end-of-life options of wood plastic composites. Witik et al. (2013) have studied the end-of-life options for glass fiber-filled polymer composites and obtained exactly the same results as in this study when comparing landfill deposition and incineration. In addition, Lazarevic et al. (2010) have reviewed studies concerning the end-of-life options for plastic wastes comprehensively, and the results obtained are in agreement with the results obtained in the present study.
5 Conclusions

The aim of this study was to study the impact of a recycled mineral filler on the various properties of wood plastic composites and to determine the critical factors affecting the formation of the properties. The environmental performance of recycled mineral wool as the filler in WPCs was also assessed and compared to glass fiber. The study revealed that recycled mineral wool can improve certain properties of WPCs. Mainly the moisture and fire resistance properties could be improved with the use of recycled mineral wool, while challenges remain in improving the mechanical properties. The environmental performance of recycled mineral wool as the filler in wood plastic composites was found to be superior compared to glass fibers.

The mechanical properties were mainly weakened by the non-optimal adhesion of recycled mineral wool fibers to polymer matrix. The strength of interfacial adhesion was a major factor especially in the formation flexural, tensile and surface hardness properties. All these properties decreased as the recycled mineral wool content in the composites increased. The impact strength was less affected by interfacial adhesion, and improvements in the impact strengths of the composites were observed when recycled mineral wool was included in the composites.

The strength of interfacial adhesion between filler particles and the polymer matrix is affected by a few different factors, the formation of chemical bonds and mechanical interlocking being the two most important ones. Chemical bonds, including both covalent and hydrogen bonding, formed between the filler particles and the polymer matrix can greatly improve the mechanical properties of composites. Based on the results of the present study, it would seem that the chemical adhesion between the recycled mineral wool fibers and the PP matrix was not optimized, and therefore the reinforcing potential of the mineral wool fibers was lost, at least partially. Similarly, due to particle morphology, the mechanical interlocking of the recycled mineral wool fibers into the polymer matrix were weaker than the interlocking of wood fibers. Recycled mineral wool fibers had extremely smooth surfaces and more uniform fiber diameter compared to wood fibers, which reduced their mechanical interlocking potential.

The use of recycled mineral wool improved the moisture resistance properties of the composites notably. Especially the water absorption and thickness swelling of the composites were significantly reduced when recycled mineral wool was added into the composites. Cyclic resistance was improved only in certain cases, but even in the worst case the performance was comparable to the situation where no recycled mineral wool was used.

The water absorption of wood plastic composites depends mainly on the wood content of the composite. The plastic fraction of the composite does not absorb water, and neither do the inert mineral fillers. Furthermore, the water absorption into wood fibers completely encapsulated in the polymer matrix is reduced compared to wood fibers exposed at the surface of the composite. The decrease of water absorption observed in the present study
was due to the replacement of hydrophilic wood fibers with hydrophobic recycled mineral wool fibers and closure of the pathways for water in the polymer matrix caused by recycled mineral wool fibers. The thickness swelling of composites is affected by both the degree of water absorption and the mechanical properties of the composites. Lower water absorption values will usually lead to lower thickness swelling. In some cases the lower mechanical properties of composites could increase the thickness swelling of the composites.

The moisture resistance properties are important in many outdoor applications of wood plastic composites, such as decking products. As recycled mineral wool can be used to improve the moisture resistance properties of composites significantly, decking products and other similar applications could be some of the most potential product applications of wood plastic composites containing recycled mineral wool.

The heat build-up of the studied composites increased as the recycled mineral wool content in the composites increased. It was found that the changes in the heat build-up of the composites were almost entirely due to changes in the color of the composites caused by the recycled mineral wool fibers. A model was presented to predict the heat build-up of the composite based on the recycled mineral wool content.

The total heat release rate and peak heat release rate were the main fire resistance properties studied. It was shown that the total heat release of the composites decreased when the content of recycled mineral wool in the composites increased. The peak heat release rate was unaffected by the addition of recycled mineral wool fibers into the composites. It was concluded that the peak heat release rates were caused by the pyrolysis of the polymer matrix. Recycled mineral wool fibers, as inert material, did not affect the pyrolysis of the polymer matrix.

When studying the spray method application of a silane coupling agent it was found that with a certain filler loading and concentration of the silane treatment solution, it was possible to achieve results comparable to the use of MAPP. However, it was concluded that in most cases the strength of interfacial adhesion between the recycled mineral wool fibers and the polymer matrix was better when MAPP was used as the coupling agent. While the strength of interfacial adhesion was the main factor affecting the mechanical properties of the composites, also the total filler content and particle morphology affected the mechanical properties of the composites.

The use of MAPP led to better water absorption properties, most likely because the wood fiber content was slightly lower in the composites where it was used. The thickness swelling of the composites was not directly dependent on their water absorption.

In the light of the results of this study, MAPP seems to be a suitable coupling agent for wood plastic composites containing recycled mineral wool. Better mechanical properties, ease of use and cost effectiveness are the main factors promoting the use of MAPP instead of γ-methacryloxypropyl trimethoxy silane.
Conclusions

The environmental performance of glass fibers, recycled mineral fibers, and recycled PP as raw materials in wood-polymer composites were assessed. The environmental impacts of incineration for energy use and landfill deposition of the composite waste were also studied. The study showed that recycled mineral wool and recycled PP can be environmentally viable raw materials for wood polymer composites. Incineration as the end-of-life option had better environmental performance in all impact categories except for global warming potential.

Recycled mineral wool had better environmental performance compared to virgin glass fiber in every impact category. The production of virgin glass fiber was a notable source of emissions in every impact category studied. As both recycled mineral wool and glass fibers are inert materials, they performed similarly in the end-of-life operations. When recycled mineral wool was compared to wood material, the environmental performance was slightly better in the global warming potential and eutrophication potential when landfill deposition was the end-of-life option, and slightly worse in the other studied impact categories. In this study, the wooden material was virgin raw material, but if the wooden material is of a recycled origin, it is possible that its environmental performance is improved.

The material re-use of recycled PP performed well in the GWP and ADP (fossil) categories. In the rest of the categories, the performance was weakened by the emissions from the replacing energy production systems. Therefore, it can be said that the total environmental performance is greatly affected by the technologies used to produce replacement energy. Only incineration and material use of PP waste were included in this study. Still, in actual waste management scenarios, some amounts of polypropylene waste is deposited at landfills. Considering the importance of energy production and the emissions related to it, it can be assumed that material use is a very competitive end-of-life option compared to the landfill deposition of PP waste.

It can be concluded on the basis of the study that recycled mineral wool is an environmentally friendly option in polymer composite applications where the properties obtainable by the use of a mineral filler are required. The environmentally optimal option for the recycling of PP obtained from the C&D waste stream must be decided case by case. The environmental performance of polymer composites is an increasingly important driver for the future use of composites, and the utilization of recycled raw materials can improve their environmental performance further.

While it was shown that many properties of WPCs can be improved with use of recycled mineral wool as the filler, the main focus in further studies should be on improving the strength of interfacial adhesion between the recycled mineral wool fibers and the polymer matrix. The functional and environmental properties of WPCs containing recycled mineral wool were presented here, but the economic aspects of the use of recycled mineral wool are another potential topic of future studies.
From the waste producer's point-of-view, material re-use is economically alluring mainly because of the potential to completely avoid or achieve savings in landfill gate fees. From the recycler's point-of-view, the recycled material can provide savings in raw material costs, if the costs of recycling are competitive with virgin raw material prices. Recycling centers can still collect gate fees from waste producers, and use it to cover the costs of recycling partly, which means that the cost structure of recycled material is very different compared to virgin raw materials. There could be economic potential in both operating a separate recycling center collecting waste from producers, separating it to recycled raw material fractions and selling the fractions to companies producing different products, or the producing companies setting up their own recycling lines and accepting the waste materials directly from the producers of waste. However, due to recycling capacities, the costs of investments related to recycling lines and the need of technical know-how in separate recycling centers may prove to be an economically effective option.

It is known that the properties and chemical composition of recycled mineral wool do not vary notably as regards the location or time of manufacture, and therefore, it can be assumed that the results obtained here can be utilized broadly. Furthermore, the study provides a framework of research methods which can be easily expanded to cover the material re-use potential of other waste fractions.

This study proved that the addition of recycled materials into wood plastic composites does not necessarily mean that all the properties of the finished product will deteriorate. Instead, it was shown that certain properties can be improved to a level where advantages over traditional wood plastic composites can be achieved. With further development, wood plastic composites containing recycled materials can achieve a significant market share.
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