

Anna Keskisaari

## **THE IMPACT OF RECYCLED RAW MATERIALS ON THE PROPERTIES OF 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 2310 at Lappeenranta University of Technology, Lappeenranta, Finland on the 27<sup>th</sup> of October, 2017, at noon.

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# Abstract

**Anna Keskisaari**

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The trend for a more sustainable style of living, the tightening legislation of waste treatment, depletion of natural resources, and the increasing amount of waste have created a good inducement for manufacturing wood-plastic composites from different recycled materials.

The main aim of this thesis was to study how different recycled raw materials affect the properties and profitability of wood-plastic composites. Materials from construction and demolition industry and mining and packaging industry were used as fillers or as strengthening fibers. In another case, plastic materials from construction and demolition industry and from municipalities were used as the matrix. Some composites manufactured from different recycled materials were selected for comparing their costs to a composite manufactured from virgin materials. Mechanical and moisture resistance tests were carried out to evaluate the properties of the composites.

The use of recycled material improved some properties and weakened some others. Construction and mining waste improved the moisture resistance, packaging waste improved the impact strength, and plastic waste decreased the price of the composite. There was no composite where all features were improved. The change of the properties was highly dependent on the materials used. The price of the composite decreased in all cases except one, when recycled raw materials were used, and even significantly when recycled plastic was used.

The results of the study showed that it is possible to use recycled raw materials as a part of wood-plastic composites. The lower price and some improved properties encourage using recycled materials, when the features of the composite are understood. Due to the tightening legislation, there will be more material available for wood-plastic composites in the future. Reusing materials in wood-plastic composites will reduce the use of virgin materials.

Keywords: wood-plastic composite, recycling, waste, properties, WPC



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I have done this study between the years 2013 and 2017 while working as a junior researcher in LUT School of Energy Systems at Lappeenranta University of Technology

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Anna Keskisaari  
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Imatra, Finland



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## Abstract

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## List of publications

This thesis is based on the following papers. The rights have been granted by the publishers to include the papers in the dissertation.

- I. Keskisaari, A., and Kärki, T. (2017). Raw material potential of recyclable materials for fiber composites: a review study. *Journal of Material Cycles and Waste Management*, 19(3), pp. 1136–1143.
- II. Keskisaari, A., Butylina, S., and Kärki, T. (2016). Use of construction and demolition wastes as mineral fillers in hybrid wood-polymer composites. *Journal of Applied Polymer Science*, 133(19), pp. 1–7.
- III. Keskisaari, A., and Kärki, T. (2017). Utilization of industrial wastes from mining and packaging industries in wood-plastic composites. *Journal of Polymers and the Environment*. (Article in Press)
- IV. Turku, I., Keskisaari, A., Kärki, T., Puurtinen, A., and Marttila, P. (2017). Characterization of wood plastic composites manufactured from recycled plastic blends. *Composite Structures*, 161, pp. 469–476.
- V. Keskisaari, A., and Kärki, T. (n.d.). The use of waste materials in wood-plastic composites and their impact on the profitability of the product. In review, *Resources, Conservation and Recycling*.

## Author's contribution

In Paper I, the author had the main responsibility of collecting the data and writing the text.

In Paper II, the author had the main responsibility of sampling, measuring, and analysing the material, as well as writing the text.

In Paper III, the author had the main responsibility of sampling, measuring, and analysing the material, as well as writing the text.

In Paper IV, the author had the responsibility of sampling, measuring, and analysing the material as well as writing the text.

In Paper V, the author had the main responsibility of planning the study, analysing the results and writing the text.



**Abbreviations**

ABS	Acrylonitrile butadiene styrene
CA	Coupling agent
CCW	Carton cutting waste
C & D	Construction and demolition
CDW	Mixed construction and demolition waste
DSC	Differential scanning calorimeter
EC	European Commission
EDS	Energy-dispersive X-ray spectroscopy
ELV	End-of-life vehicle
EU	European Union
HDPE	High-density polyethylene
LCA	Life cycle analysis
LDPE	Low-density polyethylene
LLDPE	Linear low-density polyethylene
LW	Limestone waste
MAPE	Maleated polyethylene
MAPP	Maleated polypropylene
MSW	Municipal solid waste
MW	Mineral wool
N/A	Not available
NF	Natural fiber
NFC	Natural fiber composite
PA	Polyamide

PB	Plasterboard
PBAT	Polybutyrate
PBS	Polybutylene succinate
PCL	Polycaprolactone
PE	Polyethylene
PET	Polyethylene terephthalate
PHA	Polyhydroxylalkanoates
PLA	Polylactic acid
PP	Polypropylene
PS	Polystyrene
PTT	Polytrimethylene terephthalate
PVC	Polyvinyl chloride
SEM	Scanning electron microscopy
TGA	Thermogravimetric analysis
TS	Thickness swelling
WA	Water absorption
WEEE	Waste electrical and electronic equipment
WF	Wood fiber
WPC	Wood-plastic composite

# **1 Introduction**

## **1.1 Waste**

The world is changing rapidly and we need to move with it. The decreasing amount of raw materials raises their prices and forces companies to make more sustainable choices (Crabbe 2013). The future lack of petroleum demands more sustainable choices to reduce the dependence on petroleum. With the concern on the environment, more sustainable choices have become more relevant (Kim and Pal 2011).

The world produces enormous amounts of wastes, and in some places they are well recycled, and some others not so well. The composition and amount of waste vary significantly globally. To generalize, it can be stated that the higher the living standard, the higher the amount of waste (Hoornweg et al. 2012). Hoornweg et al. (2012) have estimated the amount of waste produced by urban residents in the world. According to them, the trend is that the overall amount of waste rises all the time, and the amount of waste per person rises when the standard of living rises.

In the European Union (EU), the total amount of waste generated in 2014 was 2598 million tonnes in all 28 member states (EU28) (Eurostat 2016). Different types of waste and their share in percentages are presented in Figure 1.

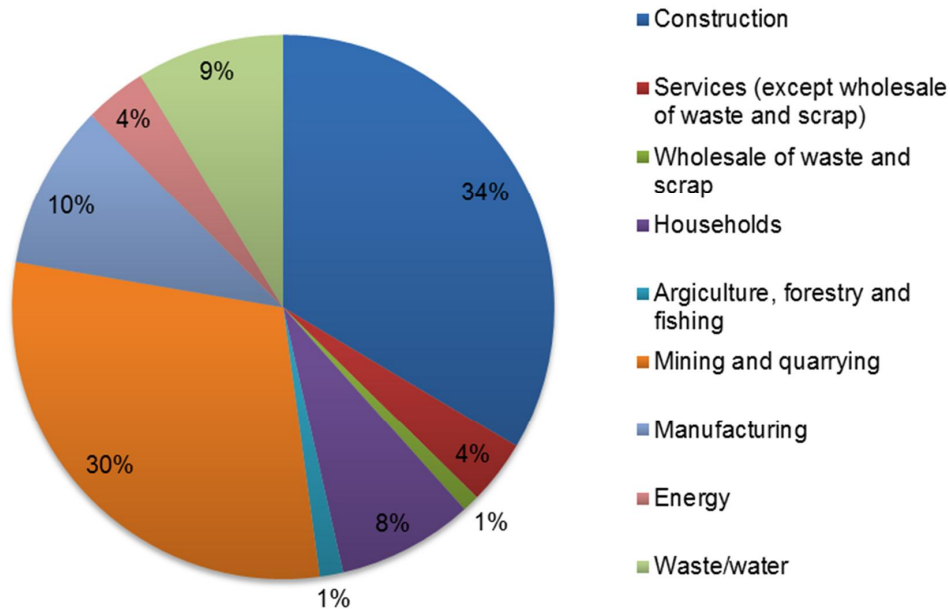


Figure 1. Composition of waste in the EU28 by economic activities and households (adopted from Eurostat 2016).

As can be seen in Figure 1, construction sites are the most significant waste producers, with mining and quarrying following closely behind. The amounts and compositions of wastes differ globally and even locally, as do recycling rates. In Europe, which may be seen as a unified area on the global scale, there are big differences in waste recycling. Eurostat has compiled statistics about different waste streams. As an example, Table 1 presents the quantity of municipal solid waste (MSW) and waste treatment in some EU28 countries.

Table 1. Municipal solid waste generation and management in selected countries in the EU28 area (Eurostat newsrelease 2014).

	<b>MSW generated</b>	<b>MSW treated, %</b>		
	Kg/ person	Recycled & Composted	Landfilled	Incinerated
<b>Bulgaria</b>	460	27	73	0
<b>Denmark</b>	668	45	3	52
<b>Estonia</b>	279	40	44	16
<b>Finland</b>	506	34	33	34
<b>Germany</b>	611	65	0	35
<b>Greece</b>	503	18	82	0
<b>Croatia</b>	391	16	85	0
<b>Cyprus</b>	663	21	79	0
<b>Latvia</b>	301	16	84	0
<b>Malta</b>	589	13	87	0
<b>Romania</b>	389	1	99	0
<b>Sweden</b>	462	47	1	52
<b>EU28</b>	492	42	34	24

Denmark produces most waste per person in Europe, 176 kg more than an average EU28 citizen. Estonia produces least, over 200 kilos less than the average EU citizen. The gap between Denmark and Estonia is 389 kilos, as much as the Romanian count. The best recycler is Germany, and the worst is Romania. 99% of Romania's waste goes to landfill, while Germany transfers no waste straight to landfill. Sweden and Denmark incinerate most wastes, and seven countries do not incinerate at all.

The numbers in the Table 1 vary considerably. The country producing most waste produces over twice as much as the lowest producing country. There are also significant differences in waste treatment between countries. The landfilling rate varies between 0% and 99%, and recycling between 1% and 65%. It must be noted that even though some countries may treat their wastes well, the amount of waste may be large. For example, Denmark incinerates more waste than Estonia ever produces. Some criticism towards the numbers in this table has to be presented, however. Have all numbers been measured in the same way, and where does the ash from incineration plants go? (Eurostat newsrelease 2014) Table 1 shows that no generalization can be made concerning waste management globally, as the practises are different in different countries. Another observation can be made: a lot of steps have to be taken to improve recycling in many countries.

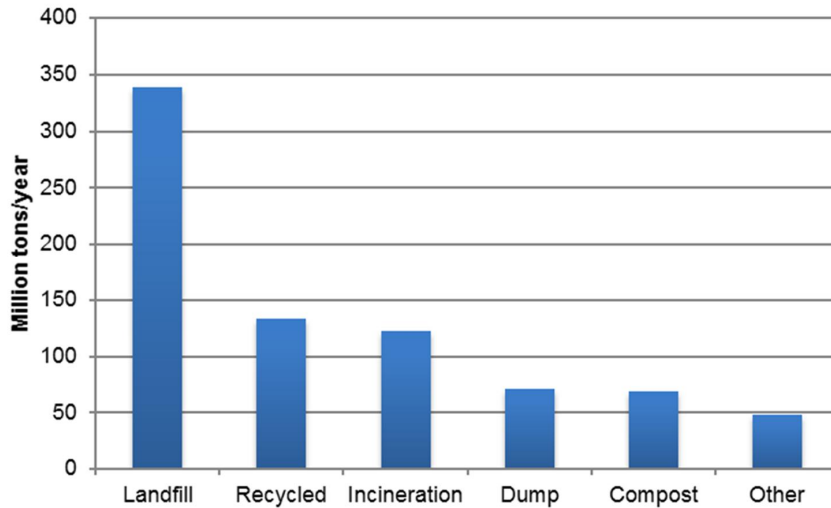


Figure 2. MSW disposal worldwide (adopted from Hoornweg and Bhada-Tata 2012).

Figure 2 expands the observation on a wider scale, as it shows MSW disposal worldwide. As can be seen, landfill is the most commonly used option. The second most common, but far behind landfill is recycling. Figure 2 shows that there is a lot of work to be done to improve waste management, but also plenty of opportunities. The figure is obviously rather simplified, as it has not been specified how for example recycling is operated. Principally the idea of the figure is to show how waste management works globally. Difference between landfill and dump is that landfill is more controlled. However, there may be differences in how different countries collect the data, so this figure has to be seen only as guiding, not the exact truth.

## 1.2 Legislation

Legislation for better waste management has been issued in the EU. Waste management in the EU is based on the waste hierarchy presented in Figure 3.



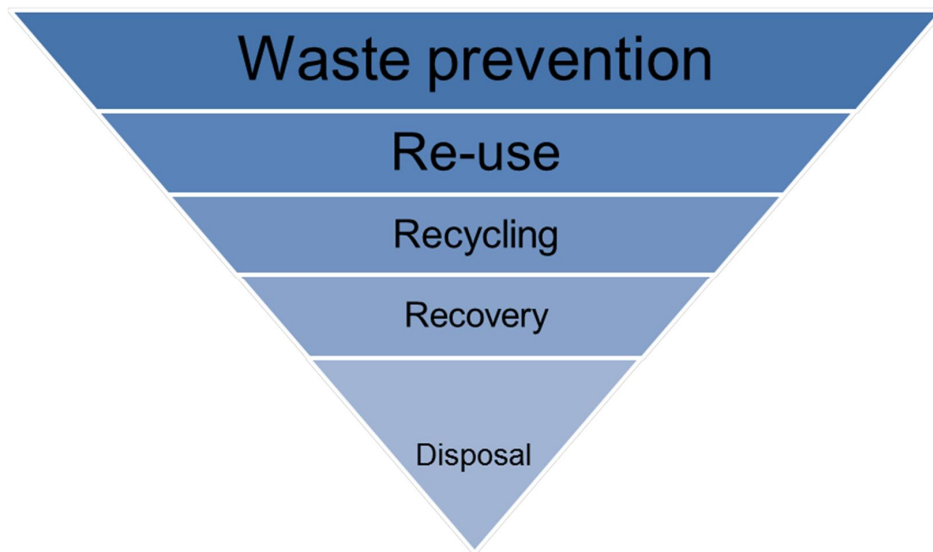


Figure 3. Waste hierarchy (adopted from European Commission 2008).

The waste hierarchy has been introduced in waste directive 2008/98/EC. The waste hierarchy is divided into five stages, as presented in Figure 3. The first step is the most desirable one, and the steps go downward with ranking merit. The first stage of the waste hierarchy is waste prevention. This stage is out of the scope of this study, and so it has not been taken into account here. The second is the reuse of material. In this stage, the materials are used as materials without major changes. The third is recycling. Stages two and three are the ones that are considered in this study. The fourth is energy recovery, and the fifth is disposal at landfill or incineration without energy recovery. (European Commission 2008)

There are several directives besides 2008/98/EC that direct waste management to a more sustainable direction. Because of the legislation, more action to reuse waste materials has to be taken. The way towards a more sustainable society will be made with different actions. Some directives order people to do something, some are more like guidance. There are also actions for easier recycling. As Table 1 shows, more sustainable waste management is already reality in some countries. There are some countries that avoid landfilling, for example. Some legislation guiding to better recycling is presented below.

The waste electrical and electronic equipment (WEEE) directive (directive 2012/19/EU) has been set to avoid hazardous materials getting to the nature, to recover valuable materials and to save resources. The directive gives people the possibility to take unused or broken WEEE-scrap to a prescribed recycling point for free.

The end-of-life vehicle (ELV) directive (directive 2000/53/EC) guides to more efficient reuse and recycling on end-of-life vehicles. The directive also guides car manufacturers to make more sustainable cars that are easier to recycle. By 2015 the reuse and recovery

of end-of-life vehicles should have been at least 95% by average weight per vehicle and year. The reuse and recycling should have been at least 85% by average weight per vehicle and year.

The packaging and packaging waste directive (directive 94/62/EC) directs towards more sustainable packaging and waste management of packaging. By 2009, 60% by weight of glass, paper and carton waste, 50% of metals, 22.5% of plastics, and 15% of wood had to be recycled. Waste combustion with energy recovery is considered as recycling.

The waste directive (directive 2008/98/EC) sets that 70% by weight of non-hazardous construction and demolition waste has to be prepared for reuse, recycling or other material recovery by 2020. By the same deadline, 50% by weight of paper, metal, plastic and glass products from households and places with similar waste flow as households has to be prepared for reuse or recycling.

### 1.3 Wood-plastic composites

Wood-plastic composites (WPC) are composites containing wood fibers or particles and a polymer as the matrix (Schwarzkopf and Burnard 2016). The term WPC is used of composites made of thermosets or thermoplastics and plant-based fibers, either wood or non-wood ones (Ashori 2008).

#### 1.3.1 Content of WPC

In WPCs, the wood is usually milled or grounded sawdust or wood flour. Besides wood, other natural fibers can be used, like jute, hemp or kenaf (Klyosov 2007). These natural fiber composites (NFCs) can be made from different parts of the plant, such as leaves, bast, fruit, seeds, or straw (Mohanty et al. 2005).

The plastic used in WPCs can be either thermosets or thermoplastics. Thermosets are plastics that can be moulded only once. They are resins like phenolics and epoxies. Thermoplastics can be heated and re-moulded several times (Ashori 2008). The most commonly used thermoplastics in WPCs are polyethylene (PE), polypropylene (PP) and polyvinylchloride (PVC), but also acrylonitrile butadiene styrene (ABS) and polystyrene (PS) have been used (Oksman et al. 2007). Besides a low price, other reasons to use these are good enough mechanical properties and low softening and melting temperatures. Plastics need to have low processing temperatures, and wood materials do not tolerate temperatures of over 200°C without damaging. Chlorinated plastics (e.g. PVC) have some restrictions in some countries due to their properties (Clemons et al. 2013).

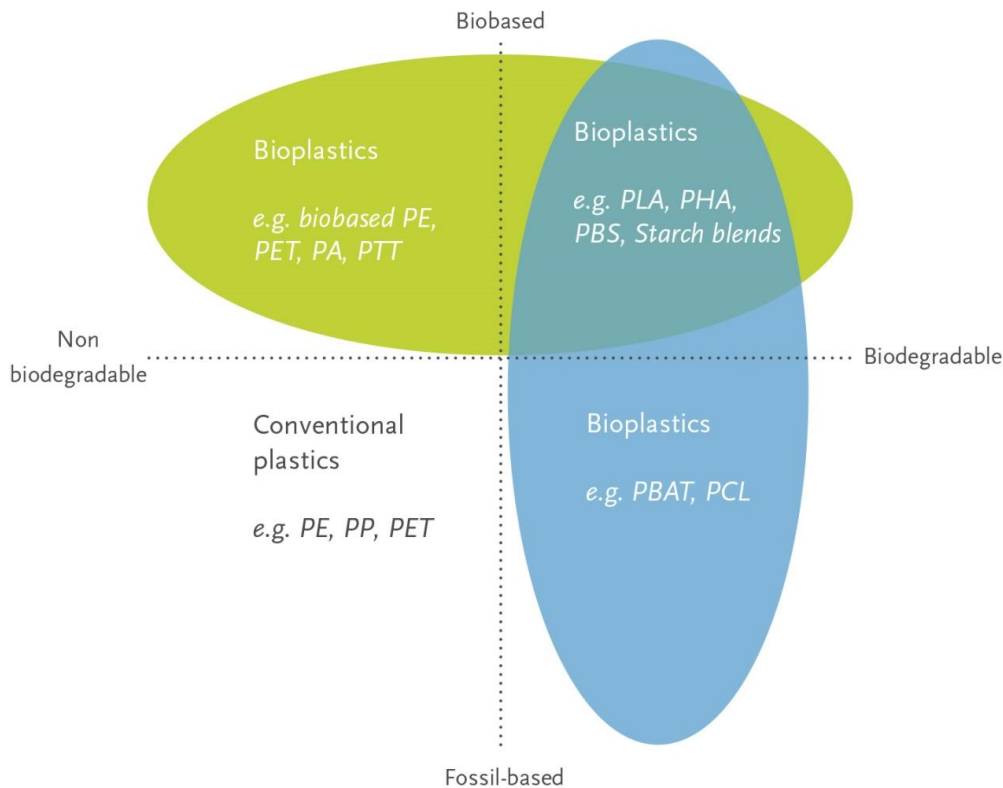


Figure 4. Classification of bioplastics. (European bioplastics 2016)

Figure 4 presents one subgroup of plastics, bioplastics. Interest in these products has increased in recent years (Aeschelmann and Carus 2015). As can be seen in Figure 4, there are different kinds of biopolymers, depending on their origin or biodegradability. Biobased plastics have been seen as one solution for the waste disposal problem with petroleum-derived plastics (Mohanty et al. 2007). Bioplastics can be biobased, biodegradable or both.

Besides strengthening fibers and plastics, WPCs consist of other materials, so-called additives. Additives are a common name to a group of different materials that are added to composites for several reasons. For example colorants, lubricants and coupling agents are classified as additives. Fillers are frequently used in the plastic industry, mostly mineral fillers. The main reason for adding fillers to plastic is decreasing its price, as plastic is usually more expensive than the filler. The filler might not have a great impact on the properties of plastics, and therefore it can be used to lower the material costs. (Strong 2006, Klyosov 2007)

Coupling agents are the most important additives. Due to the hydrophilic nature of bio-based fibers and the hydrophobic nature of plastics, adhesion between the fiber and matrix

might be poor (Ashori 2008). The coupling agent improves the adhesion between the wood material and the hydrophobic polymer (Lu et al. 2000). With polypropylene, a commonly used coupling agent is maleic anhydride polypropylene (MAPP) (Yeh and Gupta 2008).

### 1.3.2 Recycled raw materials in composites

Besides virgin materials, WPCs can contain different kinds of waste materials or by-products, and most often, recycled materials are used (Kim and Pal 2010). Almost all materials in WPCs can be from recycled sources. One case has been presented, where the composite was manufactured with materials of 95% of recycled origin (Innovationseeds 2014). There are studies of a wide range of different recycled materials in WPCs. For example, animal fibers have been used as a part of a WPC, like chicken feather fibers and silkworm silk fabric (Zhan and Wool 2011, Chen et al. 2017). Textile wastes have been used as strengthening fibers (Araújo et al. 2017), such as rice straw (Tawfik et al. 2017) and waste paper (López et al. 2012). Plastic wastes have also been used, like wastes from the automotive industry (Cholake et al. 2017) and plastic waste from electrical and electronic equipment (WEEE) (Sommerhuber et al. 2016).

### 1.3.3 WPC manufacturing

The manufacturing of a WPC is done in different phases, depending on how the WPC is planned to be manufactured. The manufacturing steps are presented in Figure 5.

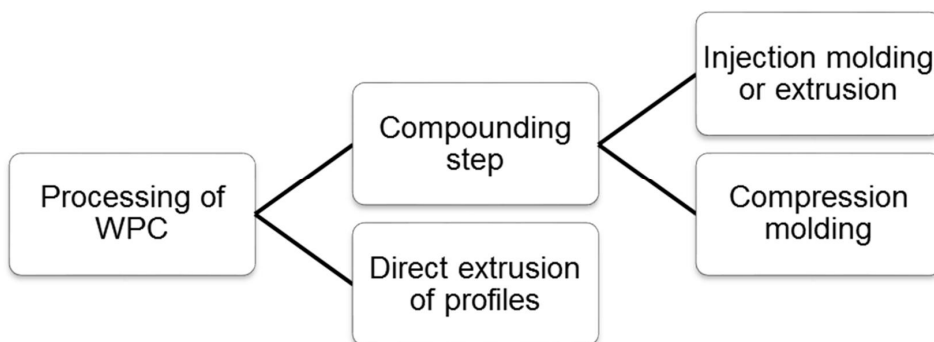


Figure 5. Processing of WPC (adopted from Oksman and Bengtsson 2007).

In the compounding step, mixing of fibers and the matrix is done before product manufacturing. This compounding step can also be made at the extruder (Oksman and Bengtsson 2007).

Two of the main manufacturing methods for WPC are injection moulding and extrusion (Kim and Pal 2011). There are two different apparatus for extrusion, a one-screw or a two-screw apparatus. There are two different applications in the two-screw equipment, counter and co-rotating screws (Kim and Pal 2011, Faruk et al. 2012). In Europe, extrusion is the most common manufacturing method for WPC, 190 000 tons of composites were extruded, while only 15 000 tons were injection moulded in 2012. Most WPCs are manufactured for the construction industry, like decking and fencing (Carus et al. 2014). In extrusion, the proportion of fibers can be as high as 80%, while in injection moulding a share of 30–40% of fiber has been used (Eder and Carus 2013). Compression moulding has been used in the automotive industry, and most of the products from NFCs in automotive industry have been manufactured with compression moulding (Carus et al. 2014).

#### 1.3.4 Properties of WPCs

A WPC is a compound of wood and plastic, and it has the properties from both materials. One of the main features is its environmentally friendly character (Godavarti 2005). The enthusiasm for using natural fibers as a part of WPCs comes from the plastic industry. Mineral fillers have been added to plastic to decrease its price. Interest in natural fibers as fillers has increased, when it has been noticed that besides the low price of natural fibers, the properties of natural fiber-filled plastic have improved (Kim and Pal 2011).

There may not always be environmental reasons behind the decision to start using natural fibers as strengtheners for plastic. Natural fibers may also have some superior properties compared to the materials used in the past. Low relative density, low cost and high specific strength are factors that have increased the attractiveness of WPC products (Faruk et al. 2012). The automotive industry has been interested in WPCs due to their mechanical and acoustic properties, recyclability, and cost and weight reduction (Godavarti 2005).

#### 1.3.5 Wood-plastic composite markets

Natural fiber composites (NFC) compete at the same markets with different kinds of products. One of the competing materials is glass fiber-reinforced composites. Joshi et al. (2004) have compared the properties of natural fiber- and glass fiber-reinforced plastics. Their research showed that natural fiber had some superior properties compared to glass fiber. Predictably, NFC had a lower environmental impact. Besides that, a lower price and lower weight of the fibers were observed. Low weight can be a useful property for example in the car industry, where there is need for lightweight materials.

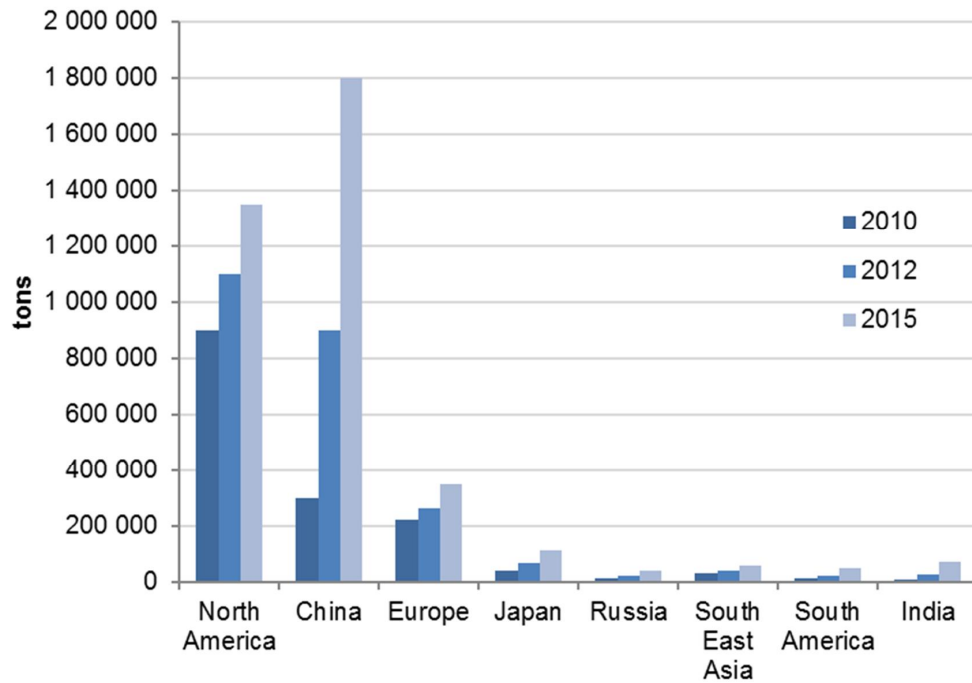


Figure 6. Production of WPCs in 2010 and 2012 and forecast to 2015 (adopted from Carus et al. 2014).

Global production of WPCs is shown in Figure 6. The markets for WPCs are growing year by year. The biggest producers are China and North America, Europe is the third, but far behind them. The other regions have only a minor production share.

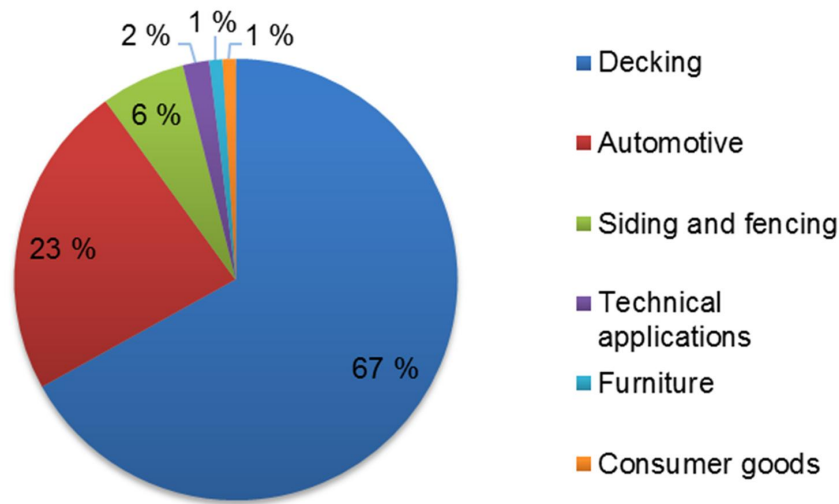


Figure 7. Production of WPCs in Europe in 2012 (adopted from Carus et al. 2014).

Figure 7 presents how WPC production was divided into different sectors in Europe in 2012. Besides WPCs, also NFCs were produced, but they have been left out of this figure (Carus et al. 2014). It can be seen in the figure that the largest application area for WPCs was decking. The second application area was the automotive industry. The rest of the sectors had no significant proportion in this area. The automotive industry had a notable share of WPC products, but the industry is a big operator in the natural fibre sector, 40% of natural based automotive products were manufactured from WPC and 60% from NFC (Carus et al. 2014).

## 1.4 Waste recycling

The increasing interest in using a wood-plastic composite (WPC) as a replacer of other products is the consequence of decreased reserves (John and Thomas 2008). One big driver for better waste utilization is the legislation. There have been several tightening legislations and regulations forcing to better waste recovery. A definition of waste has been presented in an EU directive (directive 2008/98/EC) set by the European Commission (EC). The definition is: ‘Waste is any substance or object which the holder discards or intends or is required to discard’.

Besides legislation, there are other options for better waste recycling. One demand for recycling comes from the price of material. If the production of virgin materials is more expensive than recycling, the desire for recycling materials rises. If there is a lot of raw materials available at a low price, there is no desire to recycle, as recycling may be even more expensive. Then the only way to justify the use of recycled material is its environmental values. Metal industry is one example of price-guided recycling. Mining

new material is more expensive than recycling existing material. Re-using metals is also a better choice for environmental reasons, as re-processing requires less energy than processing mined material. The greater demand for metals compared to the recycled supply keep mining operations running. The costs of mining are increasing due to that fact that the deposits are becoming more and more difficult to reach. (The Business of Mining 2012) The same kind of development has been noticed in different industries. Paper recycling has improved in recent years, and manufacturers have also economical interests in recycled materials besides the environmental aspect (EPRC 2017). It can be concluded that materials that are collected separately have to bring some advantages for the recipient. Waste electrical and electronic equipment (WEEE) is a good example, as the components may contain rare and valuable materials that are good to be recovered (Ongondo et al. 2011).

One of the points for better utilization of recycled materials is taking care of the manufacturing phase. For virgin materials, there should already be some preparation to improve the recyclability of the product. One way is considering this before manufacturing products. Eco-design and design for reuse/recycling make recycling products easier. Very homogenous materials or different materials that have been bonded together cause difficulties for recycling. Eco-design has become popular lately. Its purpose is focusing on the whole life cycle of the product. In the life of a product, all stages have some impact on the environment. Materials manufacturing, transport, use, and disposal are the main stages in the life cycle of a product. For example, in disposal, which is the point of interest in this study, less use of different materials and more recyclable solutions are encouraged. Eco-design also suggests producing more durable products. (McAloone & Bey 2014)

Figure 8 presents the EU's approach to life-cycle thinking in waste policy. With this kind of an approach it is possible to make the life cycles of products more environmentally friendly.



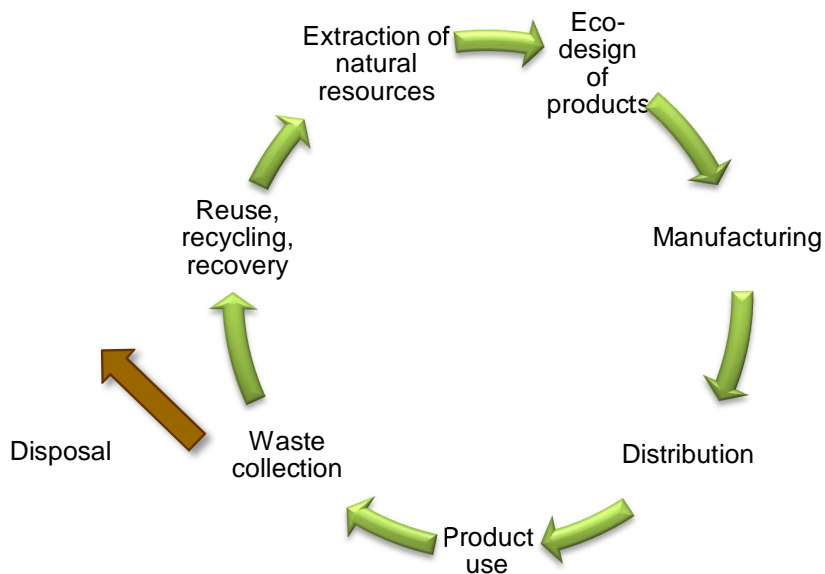


Figure 8. EU's approach to waste management (adopted from European Commission 2010).

The recycling rate of different products varies. In the United States, the Environmental Protection Agency researched the recycling rates of different products in 2010 (see Figure 9 below).

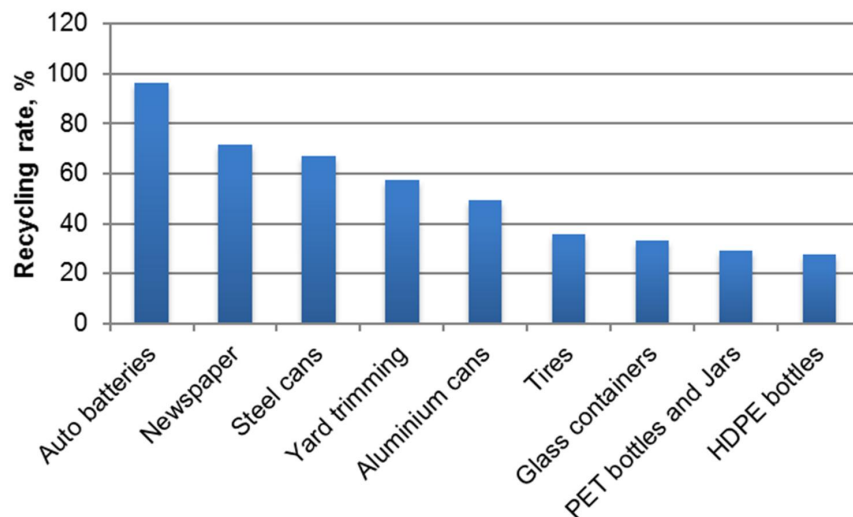


Figure 9. Recycling rates of selected products in the USA in 2010 (adopted from EPA 2011).

Figure 9 shows that auto batteries were the most commonly recycled items, while in this selection, HDPE (High-density polyethylene) bottles were the least recycled.

#### 1.4.1 Industrial waste

As Figure 1 shows, there are two large waste producers in the EU area, the construction and mining industries.

Working below the ground level is called mining and operating above the ground level is called quarrying. Both actions produce waste, but mining produces more of it. The amount of waste depends a lot on what kind of mining is in question. Diamond mining, where the concentration of the searched materials is small, generates more waste when compared to for example limestone mining, where nearly all found material can be used. (Chandrappa and Das 2012)

Industrial waste comprises a wide range of different industries, so the character of materials from this area is more extensive than for example materials from construction and demolition (C & D) industry. There are a lot of different industries producing different products. Basic industries produce products from raw materials like metal, paper, textile and plastic. Conversion and fabrication industries like packaging and automotive industries refine products from basic industries. These two industries are the greatest waste producers besides extractive industry. Some wastes are also produced by commercial activity and service industries (Chandrappa and Das 2012).

#### 1.4.2 Construction waste

As shown in Figure 1, construction waste has been found to be the biggest waste producer in the EU area. According to the EU, 25–30% of generated waste comes from the C & D industry (European Commission 2016). This amount includes a lot of exploitable waste, and the industry must begin to learn the procedure, as by 2020, 70% by weight of all non-hazardous C & D waste must be recycled in the EU (European Commission 2008).

The waste from the construction and demolition industry differs according to the type of construction. In new construction, the waste is often new materials that are leftovers or damaged. Packaging materials are also waste formed in new construction sites. Wastes from demolition sites are unclean and may contain some contaminated components. The waste is also often a mixture of different substances. (Lawson et al. 2001)

#### 1.4.3 Municipal waste

Plastic is an extremely versatile and cheap material. Because of this, it has become very popular in different applications. The increased amount of plastic used increases also the amount plastic waste (Kazemi et al. 2013). In 2014, 25.8 million tons of post-consumer plastic waste emerged. 39.5% of that amount was used for energy recovery, 29.7% was recycled and 30.8% went to landfill. There are big differences between countries, as in

some countries, most of the plastics go to landfill, while in some countries a landfill ban forces to other solutions (PlasticEurope 2016). Most of the plastic is used in the packaging industry, (PlasticEurope 2016) where the package is often used only a short time before it is thrown away (Kazemi et al. 2013).

There are several factors that affect the amount of wastes. With WEEE, there is a trend of an increasing amount of waste. Ongondo et al. (2011) found in their study that the emergence of new technologies and the fall of technology prices have created more WEEE. For example, in TV technology, the number of discarded TVs increased with the appearance of digital television. One problem with waste, especially with MSW, is its heterogeneous nature (Gundupalli et al. 2017).

## 1.5 Waste material markets

The WPC industry has grown rapidly in recent years as can be seen in Figure 6. Research in this area has been active, and a lot of different kinds of materials have been tested for their suitability for WPC composition. One of the most commonly studied issues has been different strengthening fibers. There is a huge selection of different kinds of fibers, which are usually by-products or wastes from some industrial projects.

The markets for waste materials will increase in the future, as new applications have to be found for materials that are not allowed to be put into landfill. Tightening legislation for example in the EU area forces to develop new places for recycled materials. The European Commission has worked on a circular economy package, the purpose of which is to push Europe towards a more sustainable future. One target of this package is tightening the legislation in some areas. The focus is on better waste management and encouragement for greener materials. One important aspect concerning this thesis is industrial symbiosis. The purpose is to take actions to improve industrial symbiosis, so that companies will be able to recycle their by-products and wastes for raw materials to other companies. (European Commission 2017)

As noted above, plastic is the biggest cost in the composite. Different kinds of fillers are added to the composite to reduce its price. Leão et al. (2008) disclose in their study that the differences of prices between materials are remarkable. It has been stated that the price of polypropylene is around 1.4 €/kg, whereas the prices of fibers are around 0.1–0.2 €/kg. On the basis of this, money saving might be a good inducement. There are also other benefits due to fibers, such as lightness of the material.



## 2 Aims of the study

The aim of this thesis was to study the usability of different recycled raw material fractions as a part of WPC. The work has been divided into different sections as follows:

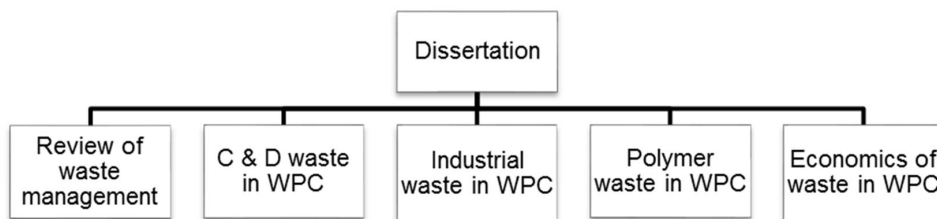


Figure 10. Structure of the thesis.

- 1) Introduction to waste as a part of WPC (Paper I)
- 2) Construction & demolition waste as a part of WPC (Paper II)
- 3) Industrial waste as a part of WPC (Paper III)
- 4) Polymer waste as a part of WPC (Paper IV)
- 5) Economical aspect of waste as a part of WPC (Paper V)

The synthesis is based on these articles. The main idea of the synthesis is to disclose how different kinds of waste materials affect the physical and economical properties of WPC. A flow chart of the structure is presented in Figure 10 above.



### 3 Materials and methods

There are five articles (I–V) included in this thesis. Their contents are presented briefly in Table 2.

Table 2. Main materials and methods used in the articles.

Article	Materials	Methods
I	A review of available waste materials as a part of WPCs	Literature review
II	Wood, PP, recycled construction waste, MAPP	Determination of mechanical properties and moisture resistance
III	Wood, PP, recycled industrial waste, MAPP	Determination of mechanical properties and moisture resistance
IV	Wood, PE, construction plastic waste, municipal plastic waste, MAPE	Determination of mechanical properties and moisture resistance
V	Materials from articles (II-IV)	Economic review of different recycled materials used in composites

The in-house manufactured research material consisted of 17 different wood-plastic composites. All materials had the same lubricant. All the materials, except the carton cutting waste (CCW) composite had the same fiber source, spruce. The composites in articles II and III had the same matrix, polypropylene (PP) Ineos Polyolefins (Eltex P HY001P) and coupling agent, maleated polypropylene (MAPP) OREVAC CA 100 (Atofina, France). In composite IV, the matrix was polyethylene (LDPE) Lupolen 2420K, LyondellBasell, Europe. The coupling agent in article IV was maleated anhydride polyethylene (MAPE) Fusabond E226. Struktol TPW 113 was used as lubricant in every article.

The waste materials differed in the different composites. In article II, plasterboard waste (PB), mineral wool waste (MW) and mixed construction waste (CDW) were used. In article III two materials were used: carton cutting waste (CCW) and limestone waste (LW). The limestone waste was mostly calcium carbonate. In article IV, construction plastic waste was used in nine composites, and one composite was manufactured with municipal mixed plastic waste.

Table 3 presents the composition of the manufactured composites with weight percentages.

Table 3. Composition of the composites used in this study.

Composite	Wood fiber	Plastic	MAPP	MAPE	Lubricant	Filler	Filler type
Ref, PP	64	30	3		3		
LW40	24	30	3		3	40	Limestone waste
LW20	44	30	3		3	20	Limestone waste
CCW		30	3		3	64	Carton cutting waste
WF-PP-MW-PB	24	30	3		3	40	Mineral wool & plasterboard
WF-PP-CDW	44	30	3		3	20	Mixed constructional waste
Ref, PE	54	40		3	3		
PB-C1	54	40*		3	3		
PB-C2	54	40*		3	3		
PB-C3	54	40*		3	3		
PB-C4	54	40*		3	3		
PB-C5	54	40*		3	3		
PB-C6	54	40*		3	3		
PB-C7	54	40*		3	3		
PB-C8	54	40*		3	3		
PB-C9	54	40*		3	3		
PB-C10	54	40*		3	3		

\*recycled

The composites in all articles were manufactured with the same equipment. For extrusion, a conical counter-rotating twin-screw extruder Weber CE 7.2 was used. The scanning electron microscope (SEM) used in articles II and III was JEOL JSM-5800 LW and in article IV Hitachi SU3500. In article IV, a Boy 30 injection molding machine was also used.

The mechanical tests were done with a Zwick Roell (Z020) testing machine. The mechanical properties were tested according to relevant standards: flexural strength was tested according to standard EN 310, impact strength according to ISO 179, hardness according to EN-1534, and tensile strength was tested with ISO 527-2. Standard EN 323



was used for density determination. For water absorption and thickness swelling, test EN 317 was used. For thermal analysis in article IV, differential scanning calorimeter (DSC) and thermogravimetric analysis (TGA) (Netzsch DSC 204 F1 Phoenix®) were used.



## 4 Results

The articles forming the basis of this thesis are presented below. The results of the studies presented in the articles are summarized in the discussion section.

### 4.1 Potential of recycled materials in WPC

The aim of Paper I was to investigate different waste streams that could be utilized as a part of a WPC. The purpose of this literature-based review was to scan the field for following research.

The amount of waste on the planet is increasing, while natural resources get more and more scarce (Hoorweg et al. 2012). On the basis of this, the recycling of waste will become more and more attractive.

There are several materials that can be utilized in wood-plastic composites. The strengthening fibers can come either from wood or some other natural fiber (NF) source. Animal fibers can be used as well (Bismarck et al. 2005, Mohanty et al. 2005). This study, concentrated on wood fibers. Plastics are another major material group used in WPCs. Both the wood fibers and plastics used in WPCs can be recycled materials.

Examination of global waste generation was made in the study. The results of the survey showed that the amount of waste and its composition varied widely in different parts of the world. The most widely used recyclable wastes came from industrial activities, construction and demolition waste, and wastes from municipalities.

Homogenous waste is easy to recycle, but heterogeneous waste can cause problems. Irregular waste requires more pre-treatment and separation. Also intermittent material flow complicates recycling. The recycling of waste materials is easier if recycling is made more attractive. Homogenous materials are easier to recycle, than for example municipal solid waste, which can contain anything.

In conclusion, the amount of waste and the rate of recycling differ greatly in different parts of the world. Thus, the same practises cannot be followed in different places. As the composition of waste differs depending on the place of origin, for WPC production, waste management has to be done locally rather than globally.

Putting waste flows into service may not be straightforward. For example, impurities, contamination, or uneven material flow must be taken into account. Despite the difficulties, it is a more desirable option to find some reasonable use for waste materials instead of disposing them in landfill.

## 4.2 Recycled construction materials in WPC

In Paper II, the potential of different construction wastes as a part of WPCs was studied. The mechanical and water absorption properties of extruded composites were studied.

Three different composites were manufactured for the tests. All composites had the same amount of PP (30%), coupling agent and lubricant (both 3%). All composites had also wood fibers, but in different amounts, depending on the amount of waste material. One composite was manufactured without a waste filler, and this was considered as a reference (WF-PP). Two of the composites were manufactured like the reference, but part of the wood fibers (WF) were replaced with construction wastes. The construction wastes were mineral wool (MW), plasterboard waste (PB) and mixed construction and demolition waste (CDW). CDW was burnt and then sieved. The purpose of burning was to get rid of organic contaminants. The composite containing besides plastic and additives 24% of WF, 20% of MW and 20% of PB was called WF-PP-MW-PB. The composite containing 44% of WF and 20% of CDW, besides plastics and additives, was called WF-PP-CDW.

Table 4. Mechanical properties of the composites.

Composite	Flexural strength (MPa)	Flexural modulus (GPa)	Charpy impact strength (kJ/m <sup>2</sup> )
WF-PP	23.35	4.44	2.07
WF-PP-MW-PB	18.75	3.83	2.34
WF-PP-CDW	17.79	4.04	3.09

The mechanical properties of the composites are shown in Table 4. Flexural properties and Charpy impact strength were tested. The flexural properties were lower compared to the reference material. As Väntsi & Kärki (2014) found in their study, replacing wood fibers with mineral wool decreased the flexural properties. Maminski et al. (2011) and Väntsi & Kärki (2014) explain the lower flexural strength of mineral wool waste composites by the low cohesion of wool and the orientation of MW fibers. Both WF-PP-MW-PB and WF-PP-CDW contained MW.

The impact strength was instead improved. This can be explained by the smaller wood content, as pure PP had higher impact strength than WPC (Nygård et al. 2008). Agglomeration of wood fibers may start crack formation.

The water absorption and thickness swelling properties improved when the wood fibers were replaced with waste fillers. The mixed construction waste composite was only slightly better than the reference material, but the composite with plasterboard and mineral filler was affected only a little by moisture. This was an expected property, as wood is the most water absorbing material, and when it is replaced with more hydrophobic material, the water absorption capacity decreases. More detailed results are presented in discussion- section.

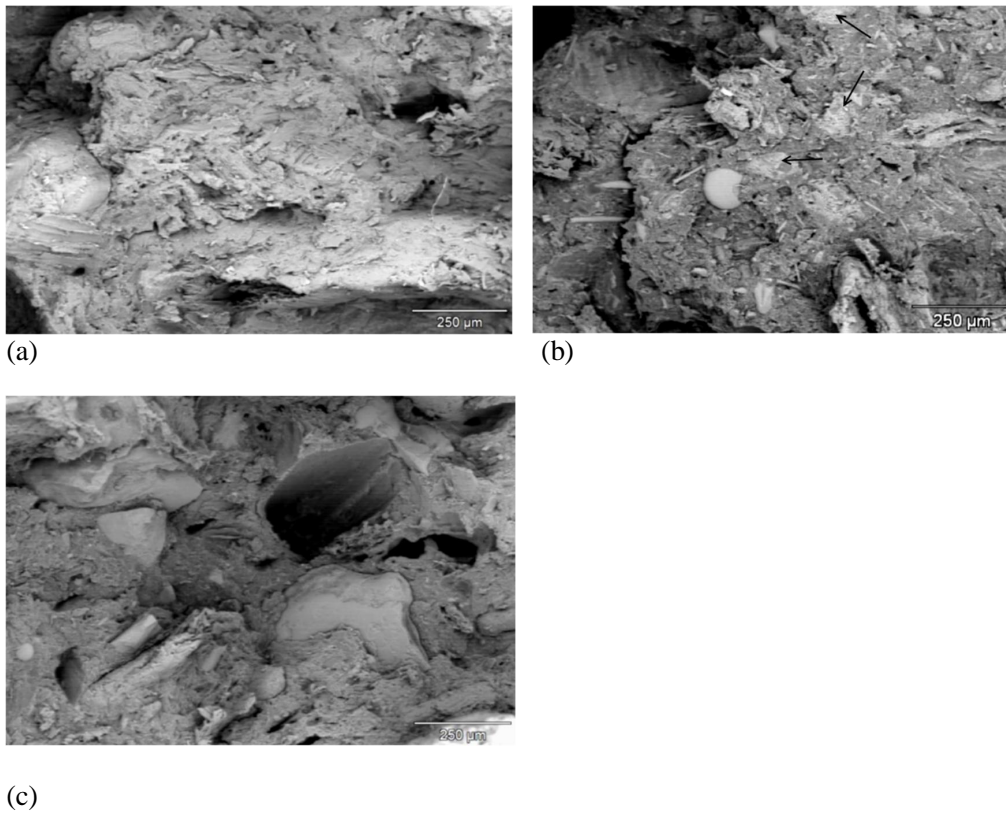


Figure 11. SEM micrograph of the composites. (a) reference; (b) WF-PP-MW-PB; (c) WF-PP-CDW

SEM was used to view the composites more closely. In Figure 11 (a) is the reference composite, (b) the composite with mineral wool (MW) and plasterboard waste (PB), and (c) the composite with mixed construction waste (CDW). The chemical composition of CDW was examined with energy-dispersive X-ray spectrometry (EDS). These studies showed that CDW contained a lot of particles in different sizes and shapes. The chemical study showed that the content of CDW was in descending order SiO, CaO, Al<sub>2</sub>O<sub>3</sub>, and SO<sub>3</sub>. The arrows in Figure 11 (b) show particles that were identified as gypsum.

The SEM study made from the fracture surface showed that the reference composite had pores, and some wood fibers were gathered together. WF-PP-MW-PB had no aggregates, and mineral wool fibers had filled the pores in the composite, which might explain the lower water absorption properties of WF-PP-MW-PB. In WF-PP-CDW, similar fibers and particles as in the WF-PP-MW-PB composite were found, so also that composite included some remains of mineral wool and plasterboard. Water absorption and thickness swelling had strong correlation to the amount of wood filler.

In conclusion, waste fillers decreased the flexural properties, but increased the impact strength. Water absorption and thickness swelling were decreased when more wood particles were replaced with waste fillers. This research showed that waste C & D fillers can be used as a part of WPC, but more careful processing would be needed to avoid the access of adverse substances to the composite.

### 4.3 Recycled industrial materials in WPC

Paper III presents the properties of WPCs manufactured with industrial wastes. Four different kinds of WPCs were manufactured Ref, LW20 LW40 and CCW. The reference composite (Ref) was manufactured without any waste materials. It contained 64% of wood material, 30% of PP, and 3% of lubricant and MAPP. Two of the composites, LW20 and LW40, were manufactured by replacing part of the strengthening fibers with limestone waste. LW40 contained 40% of limestone waste and LW20 20% of limestone waste. The fourth composite, CCW, was manufactured like the reference composite, but all wood materials were replaced with carton cutting waste. The mechanical properties (flexural strength, flexural modulus, impact strength and Brinell hardness), water absorption and thickness swelling of these extruded composites were tested. Linking between the materials in the composites was observed in the fracture surface of the composite with SEM.

First, the densities of the composites were determined. Limestone waste increased the density, while the carton cutting waste decreased it. These results were expected, as according to Huuhilo et al. (2010), mineral fillers increase the density.

Table 5. Mechanical properties of the composites.

Composite	Flexural strength (MPa)	Flexural modulus (GPa)	Charpy impact strength (kJ/m <sup>2</sup> )	Brinell hardness (N/mm <sup>2</sup> )	Density
Ref	23.35	4.44	2.07	13.1	1.05
LW40	18.90	3.58	2.21	15.40	1.3
LW20	20.44	3.77	2.56	15.77	1.21
CCW	14.03	2.42	2.88	6.66	0.99

Table 5 presents the mechanical properties of the composites. The addition of waste materials decreased all flexural properties of all composites, compared to the reference material. The biggest drop was with carton cutting waste, in which case the reduction was 40%. Limestone waste decreased the flexural properties the more of it was added. Flexural modulus was in the same level as flexural strength.

The impact strength properties were good with recycled materials, despite the character of the waste material. The reference material got the lowest impact strength results, while CCW got the best. The increase of impact strength could be explained by the fact that CCW contained a little bit of PE, which has been found to increase the impact strength

compared to PP (Dönmez Çavdar et al. 2011). Limestone waste increased the impact strength in both cases, but the results were lower with LW40, which contained more limestone waste than LW20.

Besides flexural and impact properties, also the hardness of the composite was tested with the Brinell hardness test. The limestone waste composites were the hardest, the reference being the third best. CCW got noticeably lowest results, being 49% worse than the reference material. These results were predictable, as e.g. Huuhilo et al. (2010) have noticed that calcium carbonate increases hardness, while Karimi et al. (2006) have noticed that replacing wood particles with carton ones decreases hardness.

Studies on water absorption and thickness swelling properties were also made. The limestone waste composites absorbed water clearly less than the reference material. The composite containing most limestone waste absorbed hardly any moisture. CCW had almost equal water absorption values, while thickness swelling was less than that of the reference composite. More detailed results are presented in discussion- section.

All composites were observed more closely with SEM on the fracture surface. The SEM images showed that the CCW composite had poor attachment between the fibers and the matrix. This caused poor mechanical properties, except for impact strength. The presence of PE improved the impact strength in CCW. The poor mechanical properties may have been due to the small amount of PE in the carton waste, which may have caused incompatibility between the main matrix PP. The SEM micrographs of limestone waste composites showed a smooth surface. The more limestone was added, the smoother the surface was. The smooth surface of the composite explains the good water absorption and thickness swelling properties of the composites, as there were no pores for water.

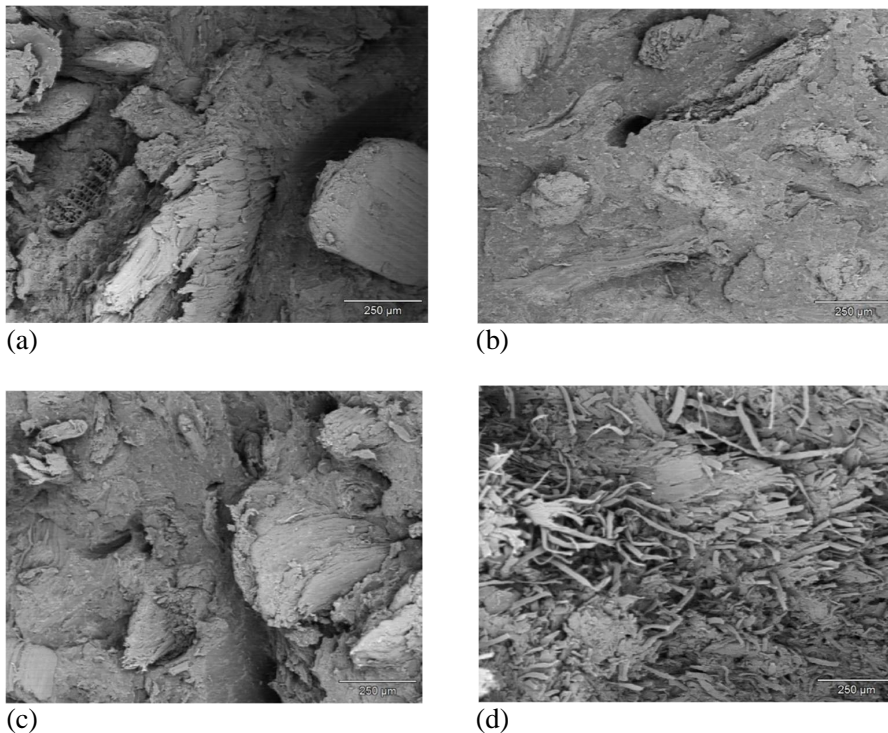


Figure 12. SEM micrographs of the composites: (a) reference; (b) LW40; (c) LW20 and (d) CCW.

In conclusion, some properties of the composites were improved. Limestone waste decreased the water absorption capacity. Hardness and impact strength were also improved, so that a smaller amount of limestone got better values than the composite with more limestone waste. Carton cutting waste improved the impact strength remarkably, and gave slightly better moisture properties, but the other mechanical properties were lower. With a better coupling, CCW might have better properties.

#### 4.4 Recycled polymer materials in WPC

Polymer wastes as a part of WPCs were studied in Paper IV. The heterogeneity of the plastic waste flow (Selke and Wichman 2004) and difficult recycling of mixed plastic waste (Shent et al. 1999, Vilaplana and Karlsson 2008) call for new recycling methods.

Eleven different composites were manufactured with extrusion. Nine of them contained waste plastic from construction sites and one contained waste plastic from municipalities. One composite was manufactured as reference material with the same amounts of the same materials as in the other composites, but the plastic used was virgin low-density polyethylene (LDPE).



The plastics to the composites were selected from different waste streams. The plastics were manually sorted, and only PE and PP were used. Most precisely, composite PB\_C9 contained flexible packaging materials and PB\_C7 consisted of hard non-packaging plastic, like pipes and furniture. Composite PB\_C10 was manufactured from mixed municipal plastic waste. Extrusion and injection moulding were used as manufacturing methods. Composites were manufactured with extrusion and plastic blends used in the composites were injection moulded.

Mechanical and wettability tests were made. The morphology was studied with a scanning electron microscope (SEM), and thermal analysis was made with differential scan calorimetry (DSC) and thermogravimetric analysis (TGA) techniques. The PE content of plastic blends was observed, and the mechanical properties were evaluated as a function of the PE content. Tensile tests were done for plastic blends a composites.

Table 6 presents the PE contents of the plastic blends. The composites PB\_C1 – PB\_C9 contained different amounts of PE and PP plastics, and composite PB\_10 contained also other plastics.

Table 6. PE content of the plastic blends.

Sample	Ref	PB_C1	PB_C2	PB_C3	PB_C4	PB_C5	PB_C6	PB_C7	PB_C8	PB_C9	PB_C10
PE%	100	53	69	72	69	64	62	32	52	84	-

Table 7 presents the mechanical properties of the composites. In the mechanical tests, the flexural and tensile strengths were lower than in the reference material, but the tensile and flexural moduli were improved. The tensile tests showed that the results got weaker after the addition WF compared to the reference material, which showed that the interfacial bonding between the matrix and WF was poor. WF had also the property of improved modulus. Interfacial adhesion did not affect the modulus properties in the same way as it affected the strength properties. For tensile strength, both a polymer blend and pure LDPE and extruded composites with a wood filler were tested. The polymer blends and LDPE samples were manufactured with injection moulding and the composites were manufactured with extrusion. All polymer blends got better tensile strength values than pure LDPE. Of the composites, the reference was better.

Table 7. Mechanical properties of the composites.

Composite	Flexural strength (MPa)	Flexural modulus (GPa)	Tensile strength (MPa)	Tensile modulus (GPa)	Impact strength (kJ/m <sup>2</sup> )	Hardness (N/mm <sup>2</sup> )
<b>Reference</b>	19.00	1.6	15.50	2.08	8.53	4.06
<b>PB_C1</b>	14.50	2.64	11.34	3.18	3.79	5.05
<b>PB_C2</b>	15.02	2.55	11.18	3.02	4.32	4.47
<b>PB_C3</b>	15.98	2.94	12.51	3.41	3.63	4.0
<b>PB_C4</b>	14.81	3.00	11.58	3.44	3.39	3.73
<b>PB_C5</b>	14.97	2.23	11.02	2.57	4.56	4.7
<b>PB_C6</b>	15.27	2.70	11.85	2.9	4.82	4.4
<b>PB_C7</b>	14.09	3.55	10.75	3.9	3.09	4.98
<b>PB_C8</b>	13.83	2.75	10.64	3.34	3.49	4.64
<b>PB_C9</b>	12.61	1.95	9.98	2.42	6.18	3.05
<b>PB_C10</b>	10.45	2.83	5.85	2.32	2.94	4.27

The impact strengths were clearly lower than that of the reference material. The hardness properties varied between the composites, but were comparable with the reference material.

The water absorption and thickness swelling were found to be better than those of the reference composite. The composites with lowest mechanical properties had also poor moisture properties. The increased wettability can be explained by poor WF/matrix properties, where pores between the matrix and wood conduct more moisture. Thermal analysis showed that PP and different grades of PE were present. The municipal plastic waste contained also other plastics. More detailed results are presented in discussion-section. SEM micrographs are presented in Figure 13.

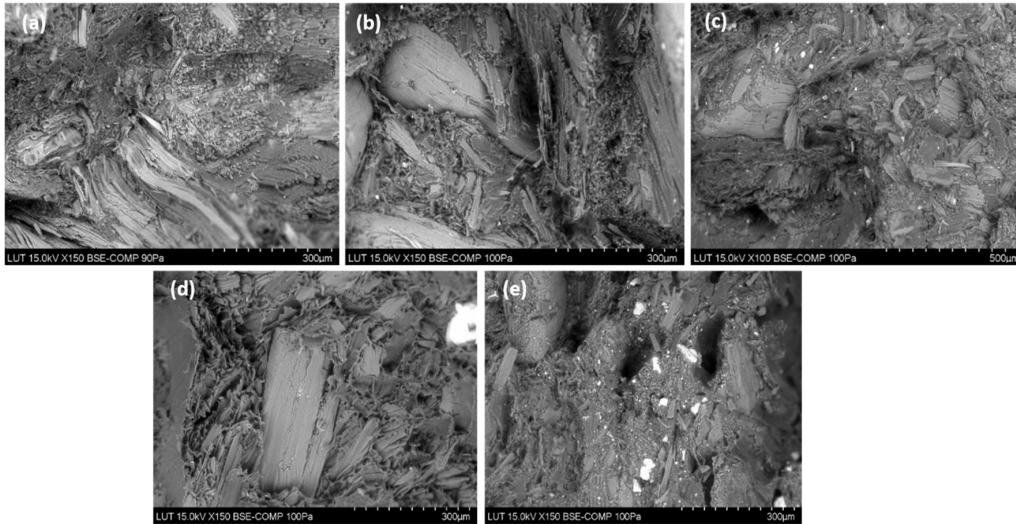


Figure 13. SEM micrographs of the composites: (a) reference; (b) PB\_C2; (c) PB\_C5; (d) PB\_C9; (e) PB-C10

The SEM micrographs show that there is a loss of adhesion in the composite manufactured from recycled plastics. In Figures 13 (b) to (e) there are some gaps and voids between the matrix and the wood particles, which are formed when wood particles are pulled out after fracture. In Figure 13 (a) the wood particles have cleaved after fracture, which can be seen as a sign of a strong adhesion between the wood and the matrix.

As a conclusion, it can be stated that no special pre-treatments were done. Adding further treatment to improve the properties might improve the performance of the material.

#### 4.5 Economical aspect of recycled materials in WPC

In Paper V, the economic consequences of using wastes as a part of WPC was studied. The purpose of this paper was to assess the impact of waste materials on the economics of WPCs. The profitability of different composites was evaluated. The composites used in the evaluation were taken from the results presented in Papers II–IV. Six composites from the previous studies were selected for the analysis.

In most studies where waste materials have been used as a part of a WPC, the different physical properties that waste materials can offer have been studied. Also life cycle assessment has been increasing recently (Väntsi and Kärki, 2015, Sommerhuber et al. 2017). However, the economic aspect received little attention in earlier studies.

In the study in Paper V, the prices for the different materials were set. The prices for the different manufacturing activities were also set. This research concentrated only in

manufacturing and material costs and all the other aspects related to the manufacturing were excluded. The purpose was to value the differences in costs when different materials are used.

The study showed that the biggest cost of the materials used in a composite was that of plastic. The total costs of the composites are presented in Figure 14. The manufacturing costs were slightly smaller than the material costs per ton in most of the cases. In manufacturing costs, the definitely biggest cost was the extrusion process. Other preliminaries before extrusion were only a small matter. The differences in manufacturing costs were low between the composites, whereas the differences in material costs between the different composites were larger. Figure 14 shows the total costs of the composites.

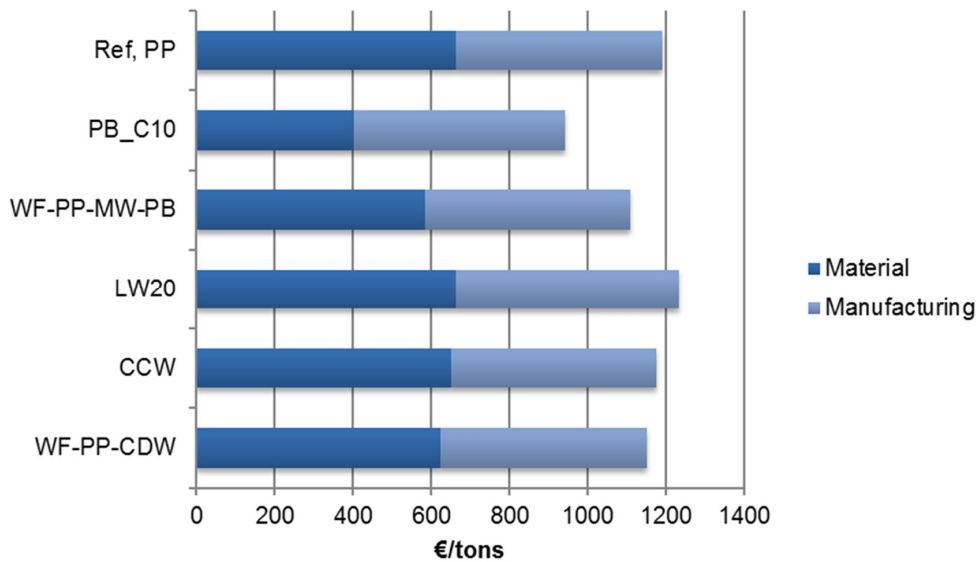


Figure 14. Total costs of the selected composites.

With individual composites, the composite that contained recycled plastic had the lowest total cost, which was due the small price of plastic. The total costs of the rest of the composites were quite alike. The biggest total cost had limestone waste. The material costs were the same with the reference, but more laborious preparation caused the higher price.

In conclusion, it can be stated that plastic is the biggest cost in the composite, meaning that waste materials do not have a remarkable effect, except for plastic waste. It has been stated previously (Kim et al. 2010) that the purpose of fillers is to decrease the price of the plastic, which was shown in this study as well. Only recycled plastic had a remarkable effect on the composite price. In most cases, the price of the composite decreased with the addition of waste materials. Waste materials did not have a remarkable effect on the manufacturing costs. In manufacturing costs, extrusion is the biggest action, and no other action affected the manufacturing costs remarkably.



## 5 Discussion

In this chapter, the results of the articles presented above are summarized. A summary of the results of the mechanical tests in the articles are presented.

The results of Papers II and III can be compared directly with each other, because the compositions of the composites were comparable. In both papers, the quantity and quality of the plastic, coupling agent and lubricant were the same. The results of Paper IV could be considered as appropriate, as there were some differences in the materials and amounts.

### 5.1 Strength properties

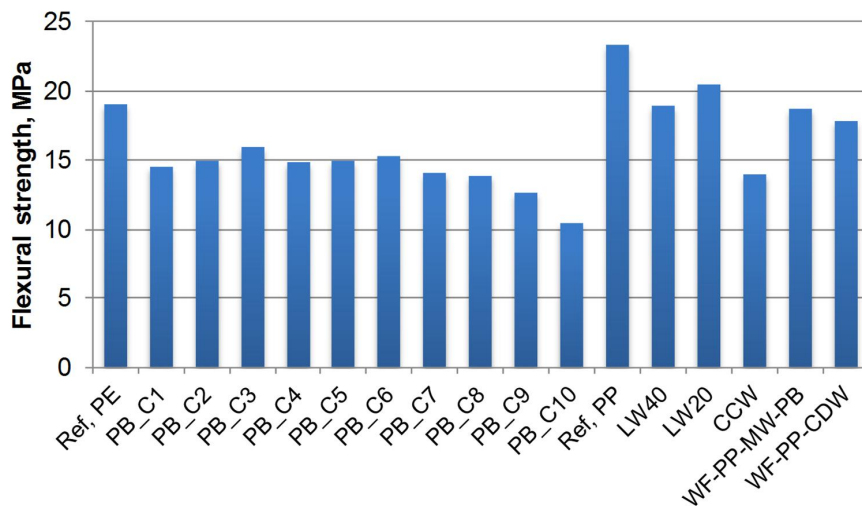


Figure 15. Flexural strengths of the composites in Papers II to IV.

Flexural strength values from Papers II, III and IV are presented in Figure 15. As can be seen, the composite Ref, PP has the best flexural strength. The second highest results get limestone waste with the lower limestone volume. Huuhilo et al. (2010) discovered that increasing calcium carbonate to the composite did not affect the flexural properties significantly. Schmaucks (2007) also discovered that increasing the amount of calcium carbonate in the composite did not increase the flexural strength. The CCW composite had the worst flexural strength in PP-based composites. This was due to poor attachment between the matrix and the fibers, which could be seen in the SEM micrographs in Figure 12 (d). Excluding CCW, there was no big difference between the other materials made from wastes studied in papers II and III.

The PE-based composites in Paper IV got mainly lower properties compared to the PP-based ones. This was due to the lower flexural properties of PE compared to PP (Strong 2006). The reference material (Ref, PE) were the best of the composites in Paper IV. This was explained with poor attachment of plastic blends and wood fibers. However, in both cases (PP & PE) the reference got the best result and the composites with different recycled raw materials were slightly weaker. The reference composite manufactured from PE got a similar strength value as the best PP composites manufactured from recycled materials. Thus, it can be said that if needed, only the strength like in the Ref, PE, PP-based composites from recycled materials is also acceptable.

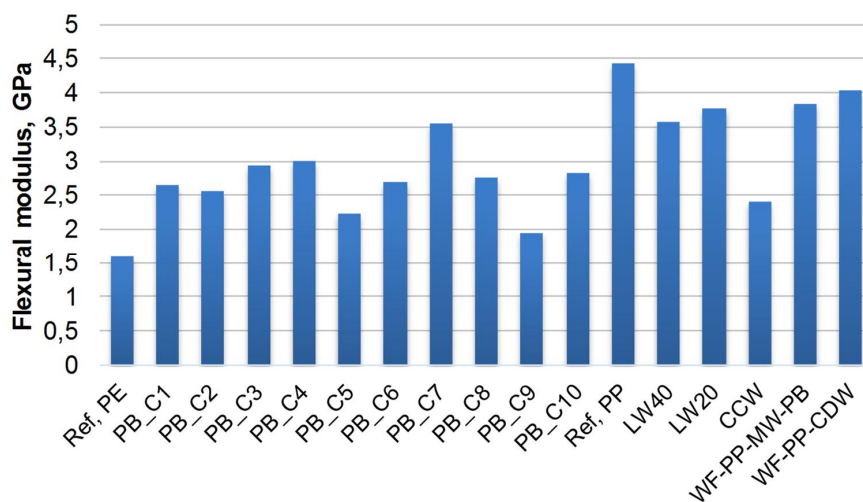


Figure 16. Flexural modulus of composites in Papers II–IV.

Figure 16 shows the flexural modulus of the composites in Papers II–IV. The flexural modulus of composites made from PP were quite well in line with the flexural strength values. For recycled plastics, the flexural modulus was improved compared to the reference material. As can be seen in the figure, the reference material from PP had a remarkably higher flexural modulus value than the Ref, PE, so it can be concluded that PP had a better flexural modulus than PE. The recycled plastic composites contained some parts of PP. The best flexural modulus in the recycled plastic group was sample PB\_C7, which had the smallest PE content. This composite, however, did not have a good value in flexural strength. This shows that interfacial adhesion does not affect the modulus as much as it affects the strength values. The same was apparent in the tensile properties.



## 5.2 Impact properties

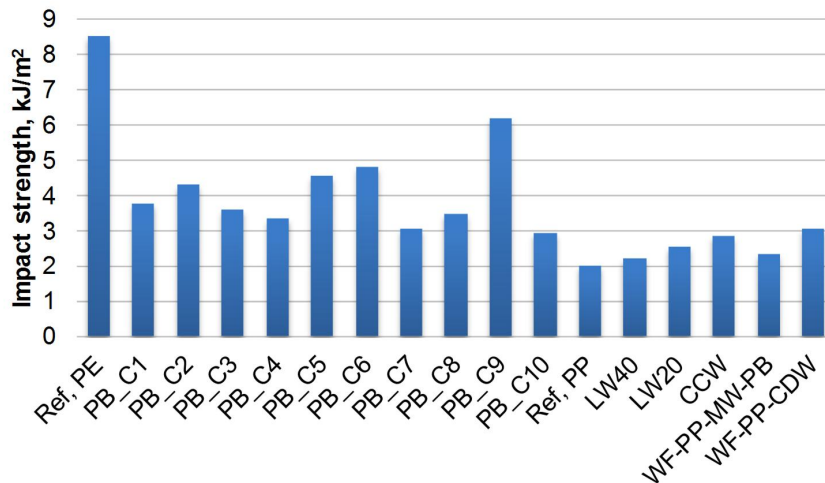


Figure 17. Impact strength of composites in Papers II–IV.

The Charpy un-notched impact strength values from Papers II, III and IV are presented in Figure 17. Clearly the best result was with the PE reference material, and the lowest with the PP reference. The high value of PE can be explained by the better impact strength properties of PE compared to PP (Strong 2006). It has to be remembered, however, that the Ref,PE contained also 10% more matrix than the Ref, PP. Composites PB-C1–PB-C10 contained some amounts of PP, so the presence of PP lowered the impact strength properties (Dönmez Çavdar et al. 2011). As can be seen in Table 6, PB\_C9 had the highest PE content. PP makes composites more fragile and rigid (Bertin and Robin 2002).

Pure PP had better impact strength values than the WPC made of the PP–matrix (Nygård et al. 2008). This can be explained by the fact that the fiber end acts as a stress concentration. Fillers and fibers also reduce the mobility of the polymer chain, which leads to a lower ability of energy absorption during the fracture. The agglomeration of wood fibers in the Ref, PP composite can have commenced crack formation and led to lower impact strength. Construction waste composites had better impact strengths, as there was not so much of bundled wood fibers.

Industrial wastes behaved in the same manner as construction waste. All recycled materials improved the impact strength compared to the Ref, PP. With CCW, the improvement was eminent. This can be explained with several factors. Two studies (Karimi et al. 2006, Migneault et al. 2014) have shown that a smaller lignin content improves the impact strength. Dönmez Çavdar (2011) has also stated that PE improves the impact strength.

### 5.3 Brinell hardness

Brinell hardness is presented in Figure 18, with the Brinell hardness properties from Papers III and IV.

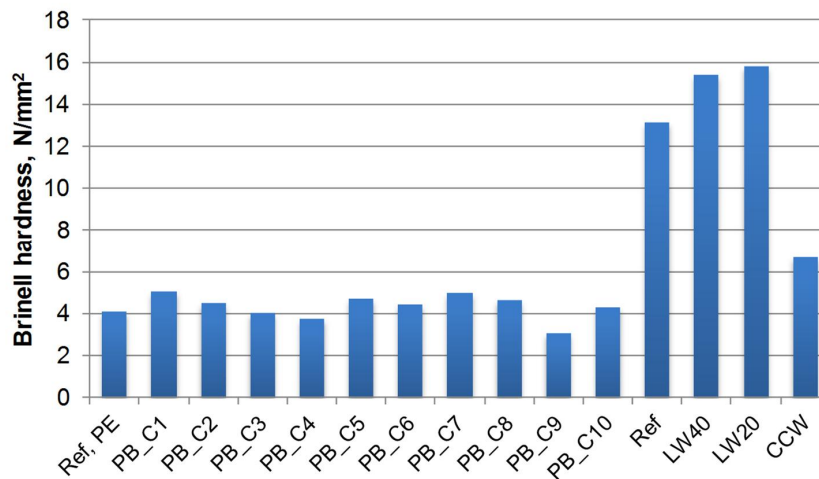


Figure 18. Brinell hardness of composites in Papers III and IV.

Huuhilo et al. (2010) have discovered that mineral fillers improve hardness. Leão et al. (2008) have found that the same type of carton material decreased the hardness of the composite. As the figure also shows, PE has lower hardness than PP. As the CCW had some amount of PE, this may have also lowered the result.

The hardness properties of the plastic blend composites varied around the reference PE. The presence of PP affected the hardness of the composite positively. PP was found to have better hardness than PE, which can be seen in Figure 18, whereas the Ref, PP had much higher value than Ref, PE. However, it must be taken into account that the Ref, PP contained 10% more wood material, and wood has been found to increase the hardness compared to polymer, so this factor also widens the gap between the references. In summary, it can be deduced on the basis of the figure that clearly better hardness is reached when using PP as a matrix.

## 5.4 Moisture properties

The moisture properties were tested with thickness swelling (TS) and water absorption (WA) tests. The tests were performed according to standard EN 317.

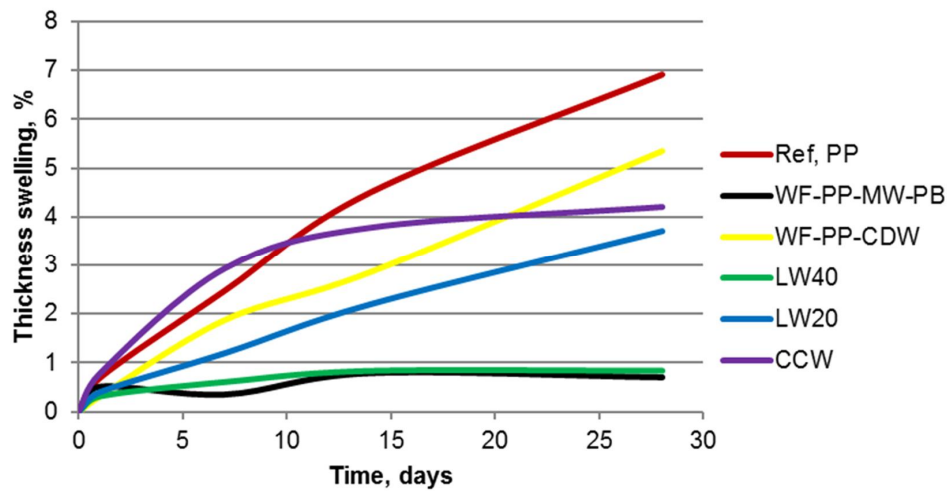


Figure 19. Thickness swelling properties of the composites in Papers II and III.

Thickness swelling from the studies in Papers II and III are presented in Figure 19. The reference composite has most swelling, while the limestone waste and mineral wool-plasterboard have the lowest TS-values. The proportion of wood fiber has a strong relation to wettability. The composites with the smallest fiber proportion got the best moisture resistance results. The CCW composite started to swell the most, but after ten days the Ref, PP passed and eventually also WF-PP-CDW did so.

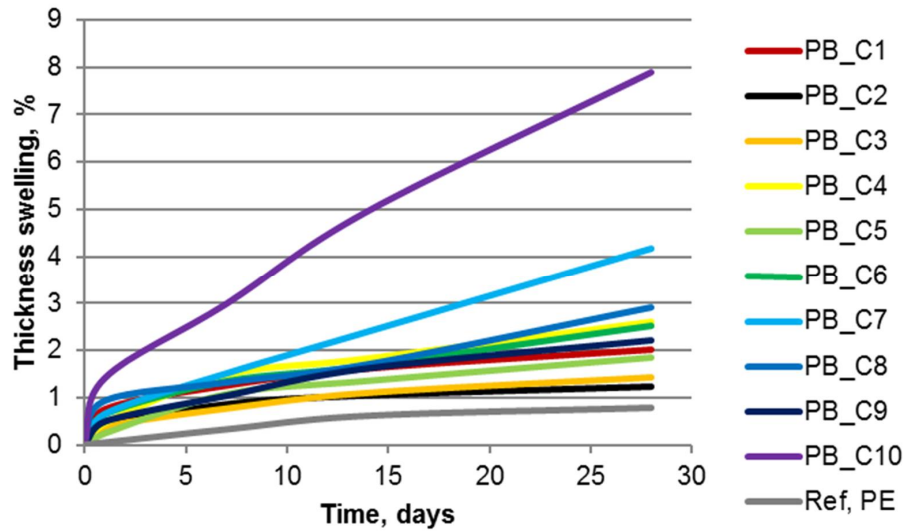


Figure 20. The thickness swelling properties of composites from Paper IV.

As can be seen in the Figure 20, the properties decreased between the plastic blends and the reference. The SEM micrographs showed that there were gaps between the fiber and the matrix in the composites manufactured from recycled polymers. This was due to poor attachment between the matrix and the fiber. The gaps allowed water to be absorbed to the composite. PB\_C10 got the highest numerical value of thickness swelling because of the worst attachment between the fiber and the matrix. All the other composites were quite alike, but the reference had the best resistance to moisture.

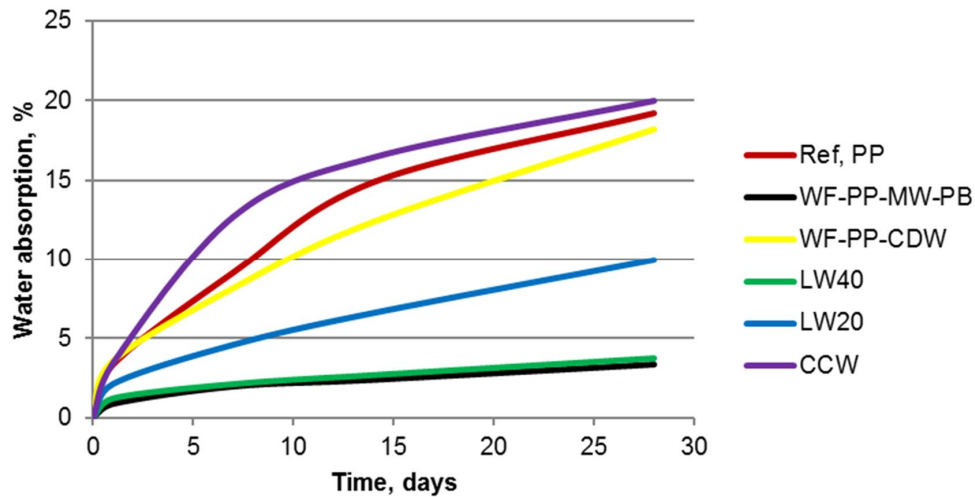


Figure 21. Water absorption properties of composites from Papers II and III.

As can be seen in Figure 21, three composites absorbed water more than the others. The most absorbing composite was carton cutting waste, the next being the reference and C & D waste. Two of the less absorbent composites were limestone waste and plasterboard-mineral wool waste. These results are quite much in line with the TS-results, although in WA the three most water absorbing composites were more similar.

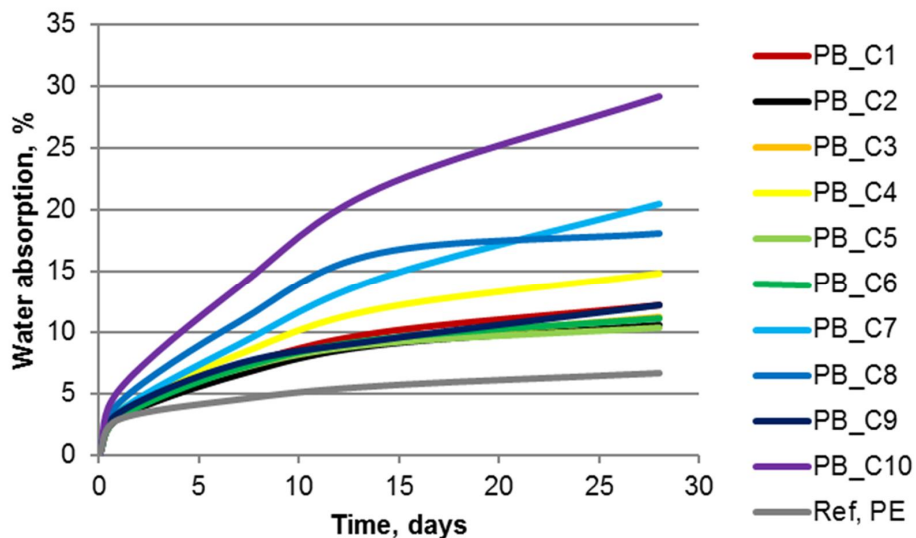


Figure 22. Water absorption properties of composites from Paper IV.

The most water absorbent composite was the one with mixed municipal plastic waste. The reference materials absorbed the least water. The water absorption properties were quite similar to thickness swelling, but in water absorption the results were not so close to each other as in thickness swelling.

Some conclusions can be drawn from the water absorption and thickness swelling results. To clarify the presentation, the data from all data was not included in the same figure. When taking all composites into account, definitely the worst properties had the PB\_C10 composite, which contained mixed municipal plastic waste. The SEM pictures showed that this composite had poor bonding between the matrix and the wood particles. The composites made from recycled plastics had more matrix materials and with less wood fiber loading, they should have been less affected by moisture. The composites made from the PP-matrix were however, filled with hydrophobic fillers, so that is why increasing the filler decreased the absorbing of moisture.

Another observation can be made of the reference materials. There was quite a big difference between the Ref, PP and Ref, PE, where PE got remarkably better values. The manufacturing methods were the same, and all other materials were the same, except the plastic and the coupling agent. There was a 10% difference in the amount of plastic, as Ref, PE contained 40% of plastic and Ref, PP 30%. Due to this, Ref, PP contained 10% more wood material. As wood is known to increase moisture absorbency, it can be seen here.

The results presented above show that the recycled plastic composites were quite equal to the industrial and constructional composites, excluding the LW40 and MW-PB composites. As in constructional and industrial composites, the moisture properties were improved, but in the recycled plastic case, the properties were weakened compared to the reference material. If the mixing ratios of different composites had been the same, the results would have been different.

## 5.5 Thermal analysis

Thermal analysis was performed by using differential scanning calorimeter (DSC) and thermogravimetric analysis (TGA). Thermal analysis was used in Paper IV. The analysis was made to the reference plastic LDPE, and to recycled plastics PB2, PB5, PB9 and PB10.

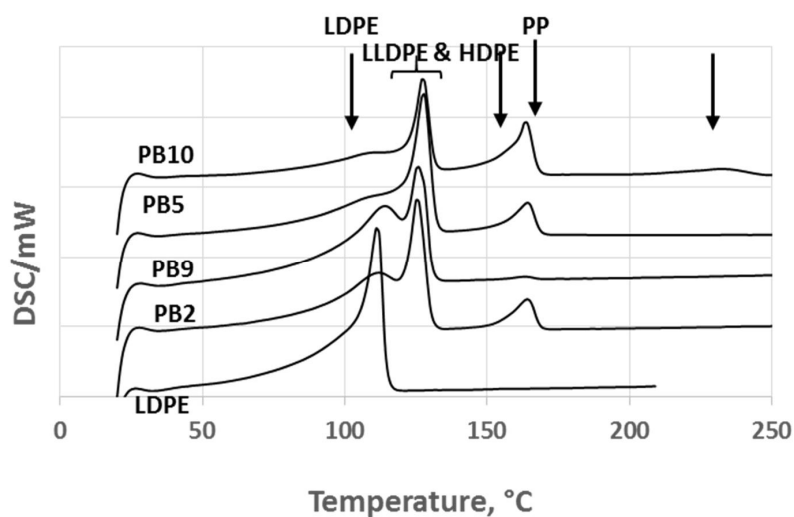


Figure 23. DSC analysis of selected plastic blends.

The DSC analysis shows the melting points which make it possible to find out which plastics there are. The peaks in Figure 23 of thermal flow curves show that three forms of PE were found: LLDPE, LDPE and HDPE. Also PP was found. In PB10 there are also additional peaks in 165°C and 235°C, which were unsigned. Household plastic waste can consist of different kinds of plastics besides PE and PP (Bertin and Robin 2002).

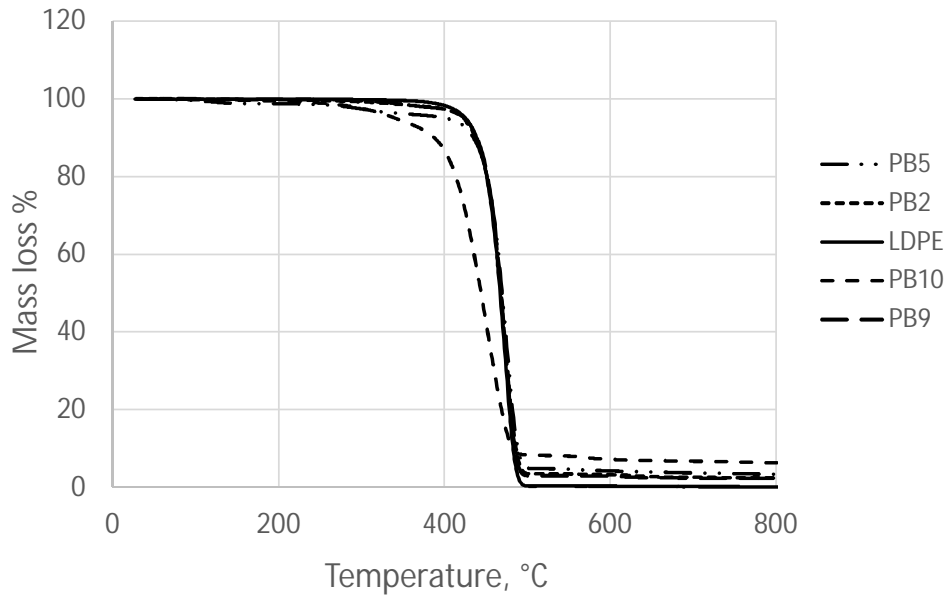


Figure 24. TGA analysis of selected polymer blends.

The mass loss rate showed a degradation process. The composites behaved quite alike, starting to degrade at the same temperatures, except for PB10 that started to degrade at 270°C. However, in every case, the degradation process started at higher temperatures than the ones used in the extrusion process.

All recycled plastics left some solid ash, while virgin PE burned entirely without any combustion products. This means that polymer blends have some non-polymeric and non-wood particles in them, like pigments and minerals or soil impurity from landfill.

## 5.6 SEM study

A SEM study was made to all composites in Papers II and III and for some of the composites in Paper IV.

In Paper II, the SEM analysis of the construction materials and the reference showed that the reference had more wood aggregates, which may have been due to a higher loading of wood. There were also some cavities in the reference sample, which were not seen in the MW-PB composite. In the MW-PB composite, the mineral wool particles had filler in the cavities. This may have led to lower WA and TS properties.

In Paper III, the CCW composite looked different from the others. This was because the fiber material was carton waste, where the material is mostly cellulose instead of whole wood fibers. Figure 10(a) shows that the attachment between the fibers and the matrix



was not good. The surface of limestone waste composite looked the smoother the more limestone it contained. The LW 40 composite looked smooth without fibers showing clearly, and this may explain the good wettability properties.

In Figure 13 from Paper IV, loss of adhesion between the matrix and wood can be seen with recycled plastics. There are gaps between the fibers and the matrix. In the reference composite, the wood particles have broken during the fracture because of strong bonding. In plastic blends, there are voids that have formed after the pull-out of wood particles.

To sum up the results, there were problems with coupling between the fiber and the matrix observed in the materials in Papers III and IV. In both papers, poor coupling was found to be caused by the presence of PE with PP. With some improvement with coupling, the results might have been better. Poor coupling was found to affect many factors, such as strength and moisture resistance.

## 5.7 Economic consequences of recycled raw materials in WPC

In Paper V, six composites were selected from Papers II, III and IV to represent different material origins.

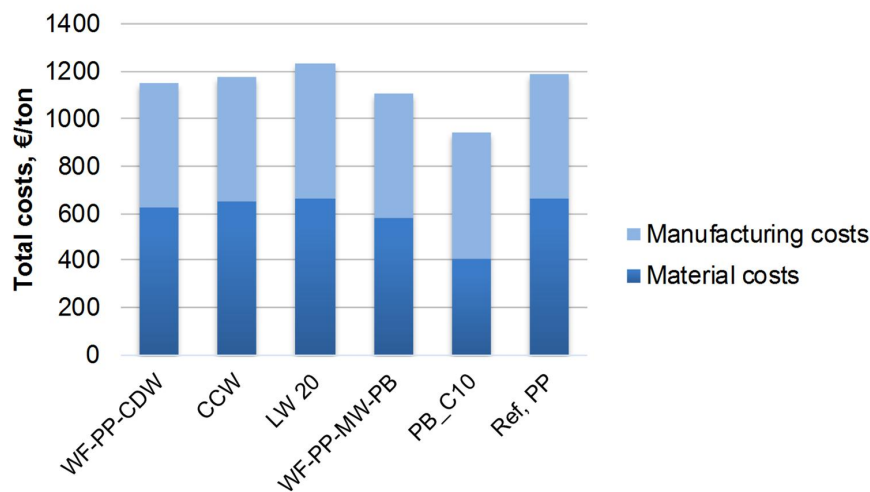


Figure 25. Total costs of the manufactured products.

Figure 25 presents the total costs for selected composites from previous studies. Compared to the reference, only the limestone waste composite was more expensive, and this was only due to more actions in manufacturing. All the others had a lower price, and especially using polymer waste lowered the price. The largest cost reduction came from the selected materials, not from manufacturing. Polymer waste had a remarkably lower material price due to the price of the recycled plastics, which was only a third of the virgin one.

The composite prices were quite similar to ones presented in the literature (Eder and Carus 2013). However, not all costs were taken into account in the present study.

All recycled materials, except recycled plastic, decreased the price of the composite only little, if at all. The material price of the wood filler is so low that replacing part of it with something else would not have a remarkable effect on the costs of the composite. The most expensive materials in the composites were the matrix, lubricant and coupling agent. The coupling agent and lubricant were added at such low concentrations, that it did not affect the material costs remarkably.

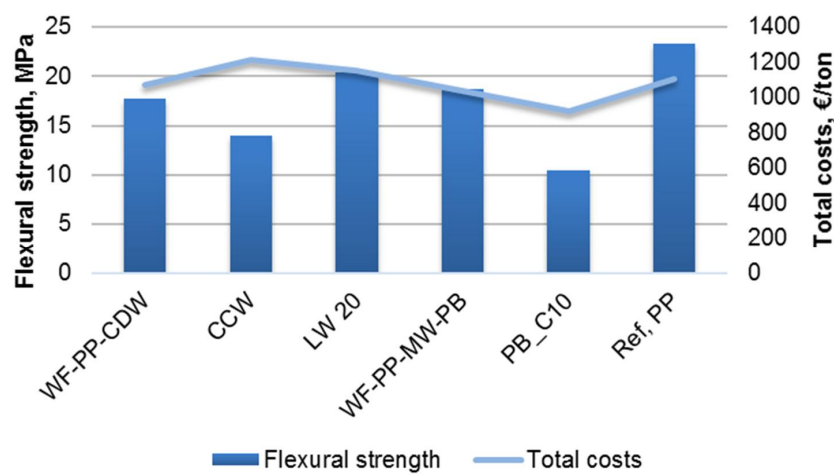


Figure 26. Flexural strength properties compared to the total costs of selected composites.

Figure 26 presents the flexural strength and cost of the composites. The closer the bar and line are in the figure, the more efficient is the composite. The reference has the best ratio between the flexural strength properties compared to the price of the composite, while CCW has the worst.

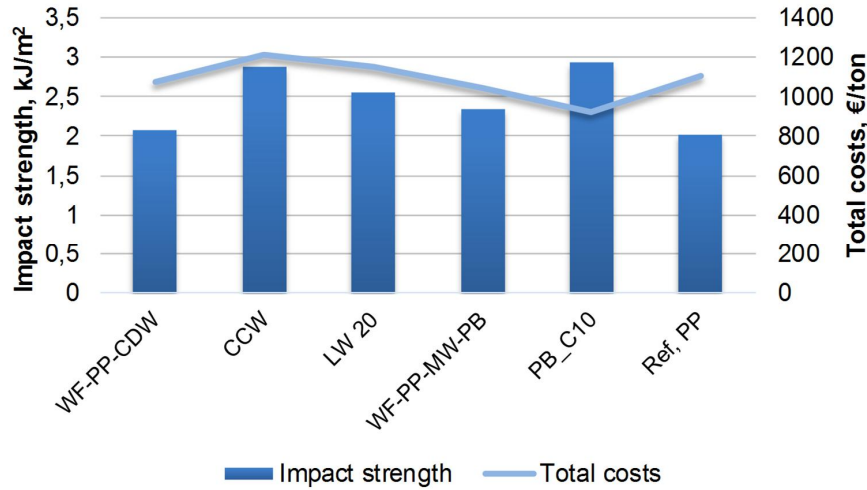


Figure 27. Impact strength properties compared to the total costs of selected composites.

The ratio between the composite price and impact strength is presented in Figure 27 above. PB\_C10 has the best ratio, while the reference PP has the worst. On the basis of this figure, if there is demand for good impact strength with the lowest possible price, a recycled plastic composite would be the optimum choice. Figures 26 and 27 have been made by valuing the properties of the composites compared to the price.

As Figures 26 and 27 show, the properties compared to composite price differ between the composites and mechanical properties. PB\_10 has the lowest value of all in flexural strength, but in impact strength it is the best. It has to be remembered, however, that the compared materials had different material ratios and different materials, so the results are not directly comparable. For example, in the case of PB\_10, the impact strength was the best of all because of the large amount of PE.

As it was found that the matrix was the most expensive material, Figure 28 shows how different amounts of matrix affected the price of the composite.

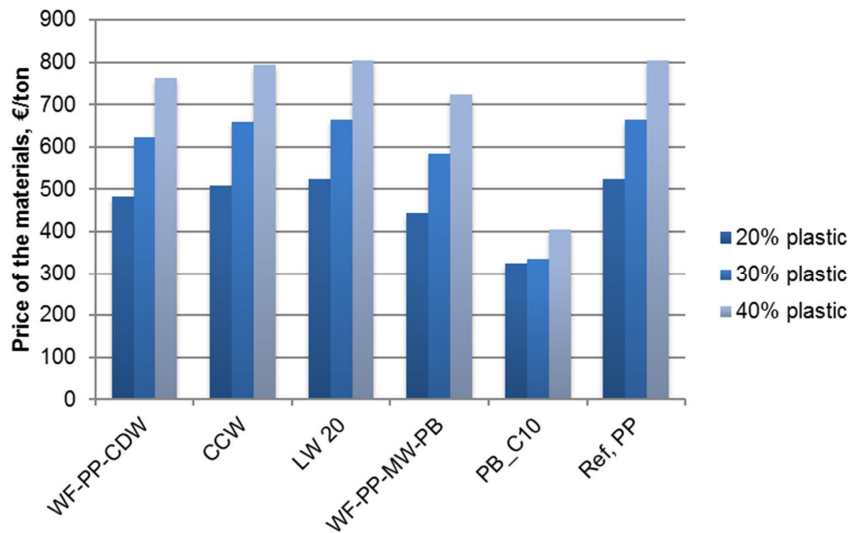


Figure 28. The effect of the share of plastic on the price of the composite material.

Figure 28 shows the effect of the share of plastic in the composite. The only composite that differs noticeably from the others is the one made from recycled plastics. This composite had the lowest price on every plastic level, and changing the share of plastic did not change the price remarkably. All other composites behaved similarly compared to each other.

As noted above, plastic had the highest effect on the composite price, while other recycled materials did not have a big impact. Figure 28 shows that changing the amount of plastic changed the price of the composite quite a lot. The price drop was approximately one third when the amount of plastic was lowered from 40% to 20% in all other composites except recycled plastic, where the change was around 20%.

## 5.8 Summary

Table 8. Impact of recycled raw materials on composites. N/A = not available.

Composite	Strength properties	Impact properties	Moisture resistance	Economical properties
Limestone waste, PP	-	+	+	0
Carton cutting waste, PP	--	++	0	0
Mineral wool-plasterboard, PP	-	++	++	+
Mixed construction waste, PP	-	++	+	+
Construction plastic waste, PE	0	-	-	N/A
Mixed plastic waste, PE	0	--	--	++

Table 8 is a summary of the properties of the manufactured composites. All composites presented in this study are not included, as some combining has been done. Construction plastic presents all nine construction plastic composites from Paper IV, as the results were quite similar. The mixed plastic waste is composite PB\_C10 from Paper IV. Both limestone waste composites were compounded from Paper III. The properties were compared to the reference materials manufactured from the same plastic (PE or PP).

The strength properties did not improve in any of the cases. However, with mixed plastic PE and construction plastic PE, the strength decreased but the modulus increased.

The impact properties of the composites from Papers II and III were improved or even remarkably improved. With the polymer waste composites, no improvement was observed, instead the results were weakened.

Recycled plastics had decreased moisture resistance while the different fillers improved it. Poor coupling between the fiber and the matrix was found the reason for the impaired moisture properties of the composites manufactured from recycled plastics. As presented in the previous chapter about moisture properties, most of the composites from recycled material got similar results. In this case, it has to be remembered, what the results are compared with, as well as the composition of the composite. The similarity was due to the different mixing ratio of the materials, as in one group of composites the results were improved, and in the other they were weakened. For this reason, the results were converged.

The most pluses got the mineral wool-plasterboard and mixed construction waste composites. In their case, the impact and moisture properties were good compared to the

reference composite. On the basis of Table 8, when putting all features together, the mineral wool-plasterboard composite would be the most preferable option, while the recycled plastic composite would be the worst.

Three physical properties against one economical property makes the latter look insignificant. That is why Table 8 has to be interpreted correctly. As noted above, all properties improved in none of the composites. If some properties were improved, some others were weakened. However, one common factor was found- the strength properties did not improve.

Table 8 shows only the development compared to the valid reference. For example, in impact strength properties, the plastic waste looks worse compared to the other composites. Still, Figure 17, shows that the recycled plastic composites got a better numerical value than the other composites presented in Table 8. In the case of impact strength, the Table 8 shows the truth, the impact strength properties decreased even dramatically in the recycled plastic case.

## 6 Conclusions

There are enormous amounts of different kinds of recycled materials in the world to be utilized in WPCs, due to the more tightening legislation and the trend toward a more sustainable future. The aim of this study was to clarify how different recycled raw materials added to WPCs influence the properties and price of composites. Mechanical properties were tested with flexural and tensile tests, as well as hardness and impact strength tests. Moisture resistance properties were tested with water absorption and thickness swelling tests. Thermal analysis was made to some composites, and SEM was used to study the morphology of the composite. In the last article of this thesis, an economical study was made to evaluate the impact of waste materials on the price of WPCs. The results showed that the type of waste materials had a prominent effect on the composite properties.

Flexural and tensile strength decreased in every situation, when some recycled raw materials were added to the composite. Flexural modulus also decreased, when fillers or fibers were replaced, but tensile and flexural modulus increased when recycled plastic was used. Recycled fillers and fibers improved the impact strength, while recycled plastic did not. Hardness was improved distinctly with the mineral filler. Cellulose fibers decreased the hardness, and with recycled polymer, the results varied quite close to the reference, depending on the material content. The water absorption and thickness swelling properties differed between the composites. All composites manufactured from plastic waste had lower moisture resistance properties than the reference material. Cellulose waste also decreased the moisture resistance, but construction and mining waste improved it; mineral waste and plasterboard and mineral wool waste even remarkably.

In profitability analysis, all recycled materials lowered the price of the composite, except for mining waste, which raised the price slightly due to more demanding manufacturing. Construction and packaging waste composites lowered the price slightly, but plastic waste lowered the price clearly.

One of the main findings was that all the composites manufactured from waste materials had some improved characteristics. No composite was found to be better than the reference in every section. As to mechanical properties, waste composites were found not to be an absolutely preferable option. However, some remarkable improvements in some properties were found. From the economic point of view, wastes lowered the price of the composite. According to the results, the price of the composite decreased when virgin materials were replaced with recycled ones.

In Paper IV, all material properties were compared to the reference manufactured from LDPE. Maybe some other kind of results would have been obtained, if one reference had been manufactured from PP, as the tested composites contained different amounts of PP. It was also noted that the presence of PP in the plastic blends improved some properties

of the composites compared to the reference made from PE. In Paper V, only one tested composite was manufactured with recycled plastics. This composite did not get the best results in Paper IV. Thus, if some other type of plastic had been chosen with better values, the recycled plastic composite might have looked preferable compared to the other composites. Poor bonding between the plastic and the wood material was found in recycled polymers. With better bonding, the results would have improved. A loss of bonding was found also in packaging waste. As a conclusion about the material properties from the different articles, it can be stated that the PE and PP – based composites worked differently. Constructional and industrial composites were quite alike compared to the polymer waste composites. No impact strength or moisture resistance properties were improved in the case of polymer waste as in the other two studies (Papers II and III).

If the application needs to have a particular property that the composite would improve, the use of the composite is acceptable. In some cases, WPCs made from recycled raw material are usable, when lower price is desired with the demanded properties. When plastic is the principal factor in the costs of the composite, all actions to replace it with cheaper materials are valuable. The use of recycled raw material has also some advantages. For decent production, there has to be steady material flow. Contamination of the materials has to be easily detectable. Heterogeneous waste materials make recycling challenging.

From the point of view of nature and circular economy, every composite manufactured from recycled materials saves the same amount of materials from virgin sources. The future of WPCs and recycled materials as a part of them looks promising. Circular economy has become trendy, and more tightening legislation requires more precise waste management. Materials are no longer seen as wastes, but more like raw material for something new. Waste producers and waste utilizers have to just find each other. Regarding the waste hierarchy, if waste prevention is impossible, the second best option for all would be effective reuse of waste. With profitable WPC products made from recycled materials, there is no point of using virgin materials. This study has shown that there are more options for waste disposal than landfill and incineration. Further studies are needed for optimizing the properties of composites.

As regards WPC materials in the future, there are some good signs. The legislation forces more tightening waste management all the time. Wastes will be seen more as raw materials to something else rather than just wastes. The waste hierarchy encourages using wastes as materials rather than in energy production. Using recycled raw materials as material prevents loss materials. If the plans of the European Union are realized, cooperation with companies in waste material markets will be improved when providers and recipients are brought more closely together.

Materials that could or have been used as a part of WPCs might become free or very affordable. When reuse, recycling, and the market are formed from waste materials, the material prices for WPC might rise. Also lack of raw materials might raise the price of materials in the future, although this concerns also other products, not just WPCs.



To improve waste recycling, recyclability should be considered already in the manufacturing phase. Heterogeneous or different materials bonded together make recycling more difficult. For example, when thinking of municipal wastes at home, food is often packaged in plastic. The plastic package may contain different plastics, for example the tray and the lid, and they are often bonded together. A carton package may have a plastic window. These things make precise recycling more difficult. Should the recycling aspect be considered already at the design phase? Development in waste separation happens naturally all the time, so should we start to improve product recyclability first or wait until the recycling technology is improved? What if there were some benefits of more sustainable design? Awareness of the state of the globe drives people towards more a sustainable direction. The amount of waste and pollution is increasing still. It remains to be seen if we will manage to lead the state of the nature to a better direction.



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## **Publication I**

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