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Faculty of Technology

Department of Energy and Environmental Technology

BH10A0300 Bachelor Seminar of Environmental Technology

**PAPERMAKING SLUDGES AND POSSIBILITIES  
OF UTILIZATION AS MATERIAL**

**Paperitehtaan jätelietteiden mahdollisuudet  
materiaalihyötykäytössä**

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Lappeenranta, 26 January 2012

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APPENDIX 1. Successful trials with paper mill effluent sludge.

## LIST OF SYMBOLS AND ABBREVIATIONS

### Abbreviations

CC	Calcium carbonate
DAF	Dissolved Air Flotation
DIP	Deinked pulp
DPS	Deinking paper sludge
DS	Dry solids
EPA	Environmental Protection Agency in the U.S.
EU	European Union
FGD	Flue gas desulphurization
GBP	Great British Pound (currency)
GCC	Grounded calcium carbonate
HW	Heavy weight
INGEDE	International Deinking Research Association
IPPC	Integrated Pollution Prevention and Control
LF	Long fiber fraction
LW	Light weight
LWA	Lightweight aggregate
MDF	Medium density fiberboard
NJDEP	New Jersey Department of Environmental Protection
OD	oven dried
PBAT	polybutelene adipate/terephthalate copolymer
PCC	Precipitated calcium carbonate
PLA	Polylactic acid
PS	Primary sludge from WWTP
RCF	Recycled fiber
SCWO	Supercritical wet oxidation
SF	Short fiber fraction
SMC	Sheet moulding compound
SPF	fiber mixture of spruce, pine and fir
SS	Secondary sludge from WWTP
U.S.	United States (of America)

UK	United Kingdom
WAO	Wet air oxidation
WW	Wastewater
WWTP	Wastewater treatment plant

Symbols

A	Annuity	[€]
N	Investment cost	[€]
r	Interest rate	[%]
T	In-service life	[a]
$Q_m$	Mass flow	[kg/a]

Chemical substances

H <sub>2</sub> O	Water
NaOH	Sodium hydroxide, lye
CaCO <sub>3</sub>	Calcium carbonate
CaSO <sub>3</sub>	Calcium sulphite
CaSO <sub>4</sub>	Calcium sulphate, gypsum
SO <sub>2</sub>	Sulfur dioxide

# 1 INTRODUCTION

The objective of this research is to identify the high solid waste streams of pulp and paper industry, emphasizing on deinking sludge, and the possibilities to utilize sludge or components of sludge as a raw material in other industries. It is also intended to gather information on the requirements that these industries set for their raw materials and if the waste streams can correspond to these requirements.

The sludge type streams of the pulp and paper industry are typically generated in water treatment and recovered paper processing. The main target is to focus on effluents of the recycled fiber (RCF) processing that have a relatively high solids content and include valuable components, like fiber and fillers. In Finland, 50 000 tons of deinking sludge, 39 000 t of fiber and coating color sludge, and 27 000 t of wastewater sludge was disposed to landfills, in 2001 (Jortama, 2003). Even though there has been research on possible reuse for these sludges, reutilization on a large scale still remains unsolved. Based on the latest research, it is intended to collect the knowledge in one report, introducing the industry fields with reuse potential and their requirements for raw materials and discuss the possibilities of the RCF by-products to comply with these requirements.

## 1.1 Background

Research to find appropriate reuse and disposal methods for waste streams is typically targeted to wastewater (WW) sludge. According to Kunzler (2001, p. 30), in the U.S., already in 1995, 26 percent of wastewater treatment residuals were recovered as energy, 12 percent used in direct land application and one percent composted. Landfilling is the predominant disposal method for sludge like rejects. Combustion of sludge is also relatively common nowadays, especially for wastewater sludge. The heat value of sludge is low or even negative, but the combustion of organics minimizes the amount of residue to dump in the landfill sites (Hynninen, 1998, p. 123). Land application and composting of sludge requires certain concentrations of nutrients to be profitable. These requirements are usually fulfilled only by sludge from biological treatment of wastewater, also known as secondary or bio sludge. Also, in many EU countries, there are restrictions for reuse of sludge in land application. (Rothwell & Éclair-Heath, 2007, p. 1)

Yet there are waste streams produced, like the deinking sludge from recycled fiber processing, that haven't been studied until recently. The use of deinking sludge in agricultural purposes has been considered to be unsuitable (Göttsching and Pakarinen, 2000, p. 537-538), which means that for conventional disposal methods, only landfilling, composting and combustion, are valid options. Göttsching and Pakarinen (2000, p. 527-528) state that in Germany, waste from three recovered paper processing mills has been successfully incinerated without exceeding emission limits. Yet the unsolved problem of ash disposal remains as landfill disposal fees rise steadily. Composting of sludge doesn't solve problems of final waste disposal either, and the sludge from recovered paper processing requires additives, which increase the expenses of sludge handling.

As the deinking sludge consists mainly of fibers, fines, fillers and coating pigments (Göttsching and Pakarinen, 2000, p. 512) it can be considered as a possible raw material feedstock for industries and products utilizing these materials. When reaching for maximum recyclability of raw materials and maximum cost-efficiency of production, at times when raw material prices are rising, it is natural to look for ways to utilize materials to the maximum. There is also pressure on paper mills to improve sludge reduction and reuse by European Commission's Integrated Pollution and Prevention Control directive, also known as IPPC (Kay M., 2003, p. 19).

Only in the last ten years has there been research on finding industry fields and applications in which materials like fiber and fillers could be reutilized. For example, as a part of Waste & Resources Action Programme, a governmental program in the UK to enhance resource efficiency, a study was made to find reuses for paper mill sludge. In this study, the possibilities of reuse in brick, cement, plasterboard and millboard production, among many others, were plotted and some trials were made (Rothwell & Éclair-Heath, 2007). These and other researches on the subject are introduced and discussed in this work.

## **1.2 Definition of boundaries**

Generation and composition of wastewater sludge is not discussed in this report, even though it is part of the term "paper mill sludge", which is often used to describe all sludge like effluents of the pulp and paper industry. This is because wastewater effluents have

been studied in various projects and methods of their generation can be found in literature. The mechanisms of sludge generation in recycled paper processing however are not that well known and therefore they will be explained in detail, for better understanding of sludge composition and reuse potential.

The generation of waste streams of recovered fiber processing is explained to provide better understanding of reject characteristics and composition. Where utilization of deinking sludge has been studied as mixed paper mill sludge, including wastewater treatment sludge or the utilization of deinking sludge requires it being mixed with combustion ash, these will be discussed respectively.

As this is research to examine the reuse potential of waste sludge from the environmental aspect, mechanical properties, such as strength or durability of fillers and fibers in RCF effluents will not be discussed in depth. Instead, the focus is on different components present in waste streams, their proportions and purity, as these set the boundary conditions for the reuse to be manageable in terms of technology.



## **2 GENERATION AND PROPERTIES OF PAPERMAKING SLUDGE**

There are diverse waste streams produced in the pulp and paper industry. By-products of a pulp and paper mill can be divided into wastewater treatment residuals (wastewater sludge), wood ash (ash produced in heat and power plant), causticizing area waste, secondary fiber rejects (postconsumer recovered fiber) and other paper mill rejects (Kunzler, 2001, p. 30). According to Bird and Talberth (2008, p. 2) papermaking discharges can be divided into residuals from wastewater treatment, ash, causticizing area waste, wood yard debris and other rejects. Sludge type discharges can be further divided into primary sludge (PS) from wastewater treatment process, deinking paper sludge (DPS) from recycled fiber processing, secondary sludge (SS) from activated sludge process and combination of PS and SS (Geng X. et al., 2007, p.346).

According to Balwaik and Raut (2011, p.300), about 300 kg of sludge is produced for each 1 ton of recycled paper. The amount of waste generated in paper production varies greatly within different regions, because of different recycling rates. In Finland, the ratio of RCF production to paper production can be expected to be smaller than e.g. in central Europe. This is because most of the paper produced in Finland is exported to other countries and therefore the amount of recovered paper is relatively low. For example, in the UK, a bit over 5 million tons of paper and board was produced in 2007 (WRAP, 2010.). Simultaneously, the production of paper mill sludge from RCF production was approximately 1 million tons (Rothwell & Éclair-Heath, 2007).

Waste sludges from a mill using secondary fiber differ from a mill using virgin materials, not only by amount but also by composition. When processing recycled fiber, a greater amount of rejects is produced because of the unrecyclable filler proportion in the raw material. This problem is especially noticeable in mills producing recycled paper from office waste, using highly filled grades as the raw material. In general, deinking mill sludge has a higher ash content whereas kraft pulp mill sludge is high on sulfur. Naturally, great variations occur within both plant types, depending on the processes and raw materials. (Glenn, 1997, p. 34.)

There are some new technologies which could possibly solve the sludge waste problem. According to Kay (2003) conversion of sludge into organic compounds usable as fuel components, pyrolysis, oxidation to produce steam or oxidation to carbon dioxide and H<sub>2</sub>O in a process called super critical water oxidation (SCWO) could be options (Kay M., 2003, p. 20.) Yet all of these processes are under development and it can take years for them to be implemented on a large scale. Also it seems unlikely that a sludge containing numerous challenging substances, such as stickies and plastics, and high ash content would be the first one to study. These options are still good to bear in mind, as the processes will be adopted in few years' time and there is a possibility to put up a pilot plant to test their potential in sludge elimination.

## **2.1 Waste generation in recovered fiber processing**

In this chapter we will take a closer look at the rejects from paper mills concentrating on mills using RCF in paper production. Even though the emphasis of this chapter is to understand the mechanisms of waste production in stock preparation and paper machine stages, it is to remember that also RCF paper mills produce conventional wastes, such as combustion ash and sludge from WWTP.

Generation of wastewater and solid wastes varies greatly between different RCF processes as it does with processes using virgin fiber as raw material. Production of packaging paper generates the least rejects, as the water flow from the WWTP is below 4 cubic meters to a ton of paper produced. As dry content, the solid waste production is below 100 kilograms to a ton paper with organic content of about 75 %. Production of tissue and market DIP generates a water flow up to 16 m<sup>3</sup>/t<sub>paper</sub>. The amount of solid wastes as dry content may be 600 kg/t<sub>paper</sub> with organic content of 40 to 50 %. As a comparison, the production of wood-containing LWC generates average WW flow of 14 m<sup>3</sup>/t<sub>paper</sub> and 46 kg/t<sub>paper</sub> of rejects and sludges. (IPPC, 2001, p. 174, 230.)

Recycled fiber (RCF) processing may be divided into two categories. The first type includes only mechanical cleaning and can contribute to products such as testliner, corrugating medium, board and carton board. The second type includes both mechanical and chemical unit operations, i.e. the deinking process. RCF process with deinking

provides pulp suitable for newsprint, tissue, copy paper, magazine grades, carton board and market deinked pulp also known as market DIP. (IPPC, 2001, p.11.)

Recycler paper processing produces water emissions, solid waste, and emissions to atmosphere. Solid waste is produced in biggest quantities in the wash deinking process, which is common in tissue production. Emissions to atmosphere are mainly due to energy production. Solid or sludge like rejects are generated in various steps of the stock preparation process, as can be seen in Figure 1 (IPPC, 2001, p. 219, 220.) Re-circulation of process water is represented by the dashed line.

Rejects from the pulper disposal system, which is usually a screen plate type equipment to collect large particles, consist mainly of contaminants like sand, metal, plastic bags, strings and wet strength paper. These are disposed of by landfilling. After the disintegration of paper, i.e. in the cleaning and screening stages, the fiber content in the rejects rises. The high density cleaning equipment is usually a hydrocyclone to separate heavy weight (HW) particles from the pulp slurry and its reject consists of glass, paper clips, textiles, staples etc. In the screening step of the stock preparation process, the portion of fiber in the rejects already exceeds 35 percent. (Göttsching & Pakarinen eds., 2000, p.510-512.)

Fractionation is one type of screening process designed to divide the fiber mixture according to a defined criteria, e.g. size or flexibility (Göttsching & Pakarinen eds., 2000, p.118). Its primary goal is to enrich long fiber fraction (LF) and short fiber fraction (SF) in two separate outlet flows for further preparation steps.

In Figure 1, a recovered fiber without deinking, is represented. Production of paper grades with high brightness and cleanliness requirements demand better purification of the pulp, and the required properties can often be accomplished by including the deinking process to RCF preparation. Also, a reduction of stickies can be achieved with deinking. (IPPC, 2001, p. 220.)

For the deinking step, process additives like NaOH, sodium silicate, hydrogen peroxide and fatty acids are applied in the pulping stage. During the mechanical cleaning and preparation of the pulp slurry, ink particles and adhesive components are dispersed and

detached from the fibers. Residual dirt specks and stickies become smaller or floatable. In the deinking, dispersed particles are then separated from the slurry by flotation, often in several stages. The average size of an ink particle is about 0,02 – 0,1  $\mu\text{m}$ , but to guarantee efficient flotation achieved with particle size of 10 to 250  $\mu\text{m}$ , ink particles must be grouped into agglomerates. (IPPC, 2001, p. 221; Götttsching & Pakarinen eds., 2000, p. 93, 153.)

The flotation process is called froth flotation. It differs from microflotation and dissolved air flotation (DAF) by selectiveness, i.e. it aims to remove only ink particles, not all solids. As the separation criteria is the different surface wettability characteristics of particles, hydrophobic particles like printing ink, stickies, fillers, coating pigments and binders get separated from fibers and end up in the reject. (Götttsching & Pakarinen eds., 2000, p. 151.)

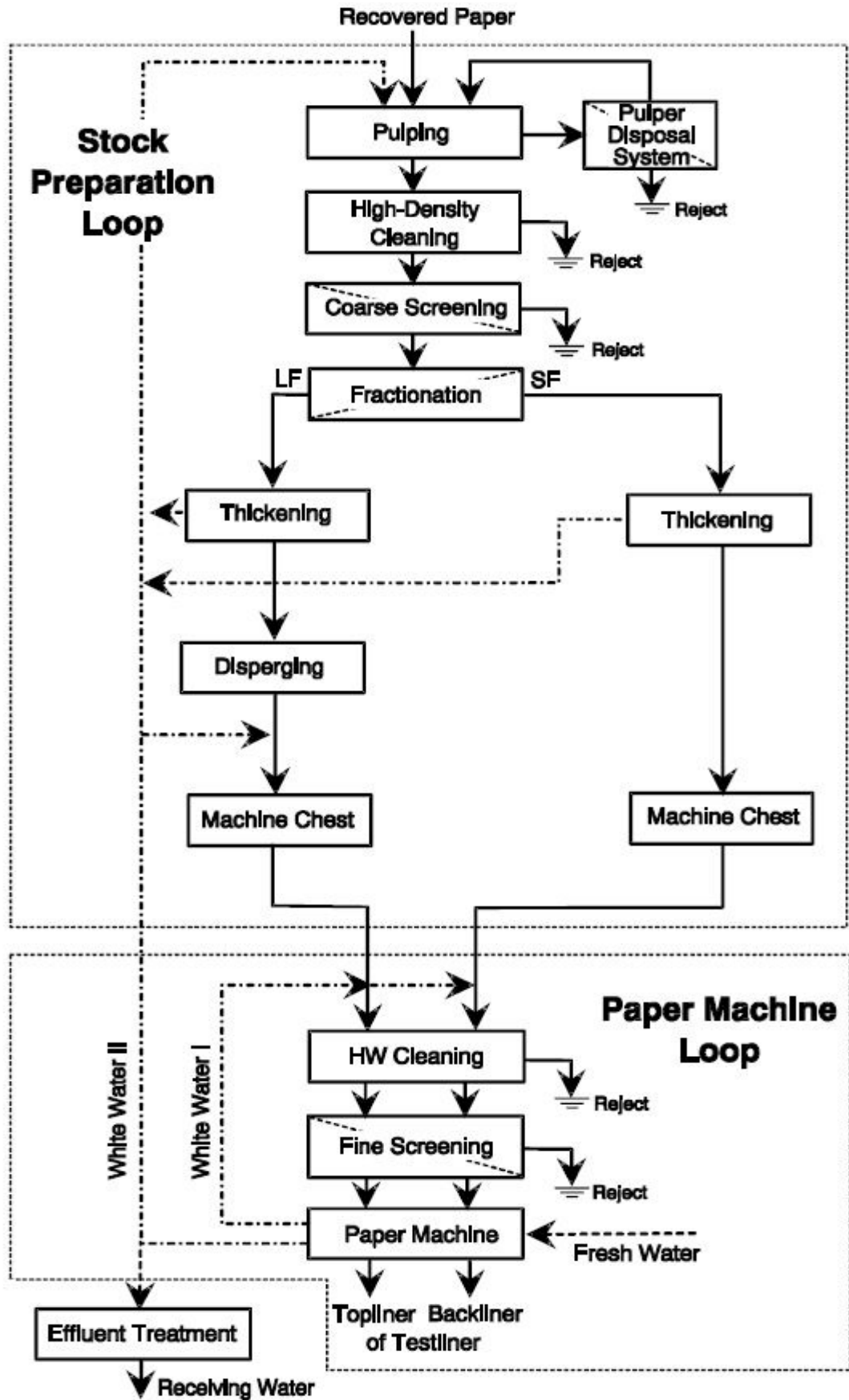
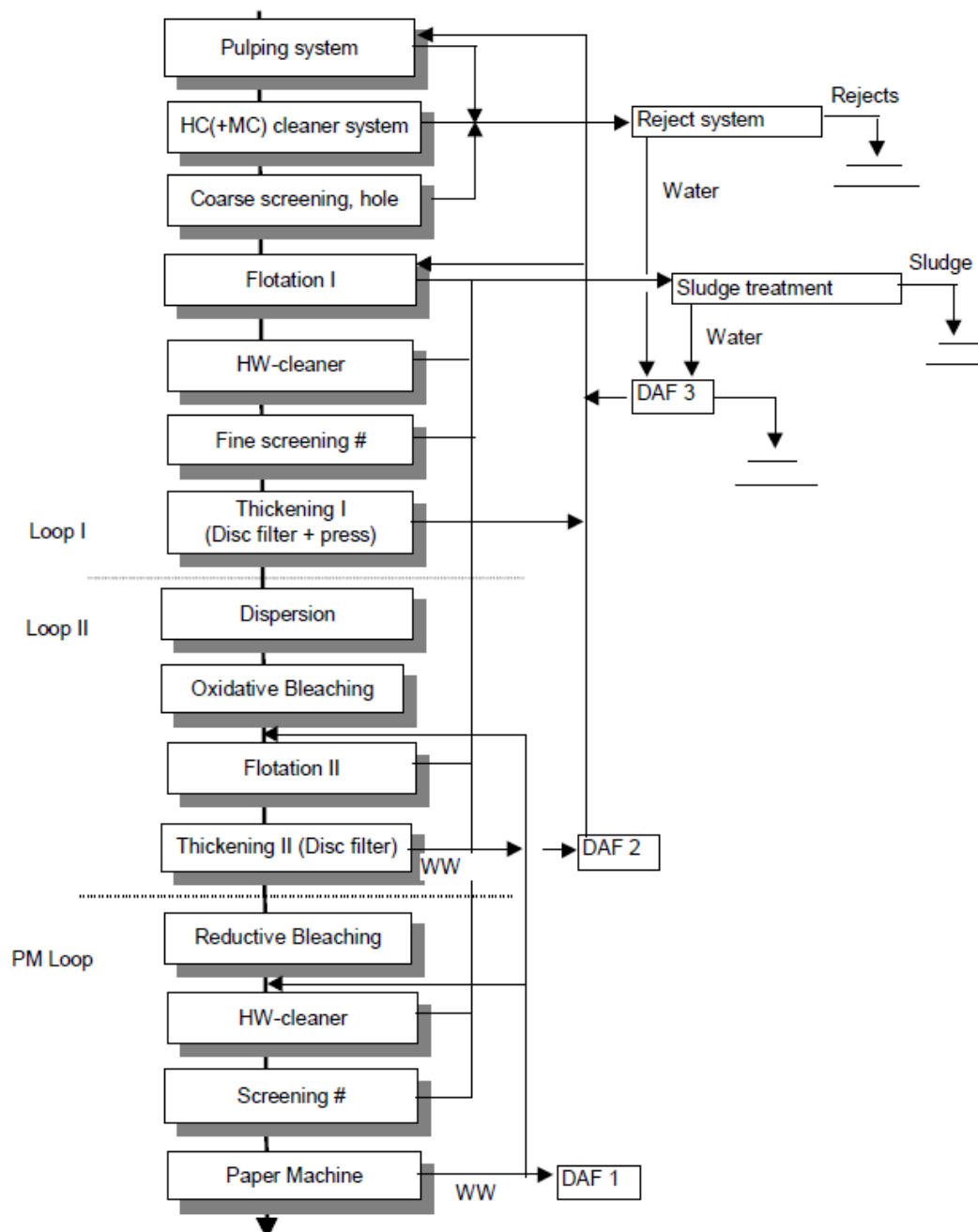


Figure 1. Flowsheet of a typical plant to produce 2-ply testliner from recovered paper (IPPC,2001, p.220).

A flow chart of production of improved newsprint from recovered paper, with reject handling and water circulation is represented in Figure 2. It can be seen that the solid rejects, with high level of contaminants, from mechanical preparation have a rejects system of their own. The generated sludge, collected at the beginning in the first flotation step and continuing all the way to the last screening in paper mill loop, is discharged to the sludge treatment process. The sludge treatment process aims to dewater the sludge to a smaller quantity and to re-circulate the process water back into the process. The sludge is disposed after dewatering. The froth and other rejects from the deinking process are separately dewatered with a centrifuge or a wire press up to 50% DS (IPPC, 2001, p. 221). As most of the valuable substances are removed in flotation and latter process steps, the rejects fed to the sludge treatment in Figure 2 together contribute to the deinking sludge with best reuse prospects.

Another possibility for ink, filler and stickies removal is a process called wash deinking. It means a specific type of dewatering process, which removes fines and fillers smaller than 30  $\mu\text{m}$ . Wash deinking is more commonly used in the U.S. than in Europe. The washing aims to remove fillers and coating particles, fines, micro stickies and ink, combined with simultaneous removal of dissolved and colloidal contaminants from the process filtrate. (Göttsching & Pakarinen eds., 2000, p. 176.) Often in the production of very clean RCF pulp, e.g. for LWC papers, flotation and wash deinking follow each other in the process loops while they complement each other (IPPC, 2001, p. 221).

Production of high quality tissue paper and market DIP requires that aside from coarse contaminants, also inks, stickies, fines and fillers have to be removed. The greatest difference between newsprint production and tissue production is a step called de-ashing which means the removal of fines and fillers. To produce the same amount of pulp, 30 to 100 percent more recovered paper is needed. (IPPC, 2001, p. 225.) This means that the amount of waste produced in the process increases. This is one of the reasons why the pulp and paper industry is looking for ways to reuse the sludge. If a profitable utilization method for these sludges were established, they would not only be able to profit from the waste but also to produce better quality (more valuable) products at a lower cost.



**Figure 2.** Lay out of a RCF process to produce newsprint (IPPC, 2001, 224).

## 2.2 Composition of sludge

Paper mills that use recycled fiber usually generate more solid waste than the ones using virgin fiber. Residue from mills using RCF consists almost entirely of inorganic fillers, coatings and short paper fibers washed out in the process of fiber cleaning. (Bird & Talberth, 2008, p. 2-3.) Wiegand and Unwin (1994, p.92) state that recycled paperboard

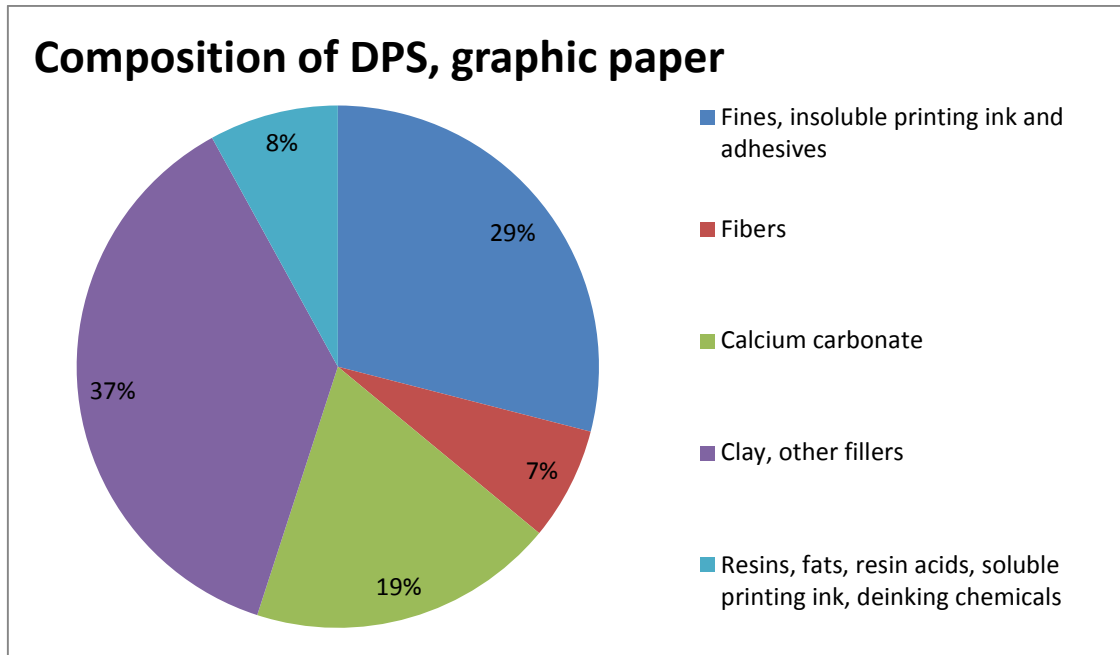
mills usually recycle their primary sludge back to the fiber processing system. This may explain partially the low fiber content of recycled fiber utilization processes.

Rejects from the pulper equipment of a mill using raw material fractions of 60% old newspaper and 40% old magazines had a dry content of 70%, of which 52% plastics, 27% flakes and fibers (originating mostly from wet strength paper clusters), 7% of each wood, metal and textiles. The amount of this reject was from 0,7 to 1 percent of the amount of air dried recovered paper. (Göttsching & Pakarinen eds., 2000, p.511.) Due to the high level of contaminants, low proportion of valuable substances and relatively small production amount, it doesn't seem reasonable to find reuse purposes for fiber or filler fraction of this reject stream.

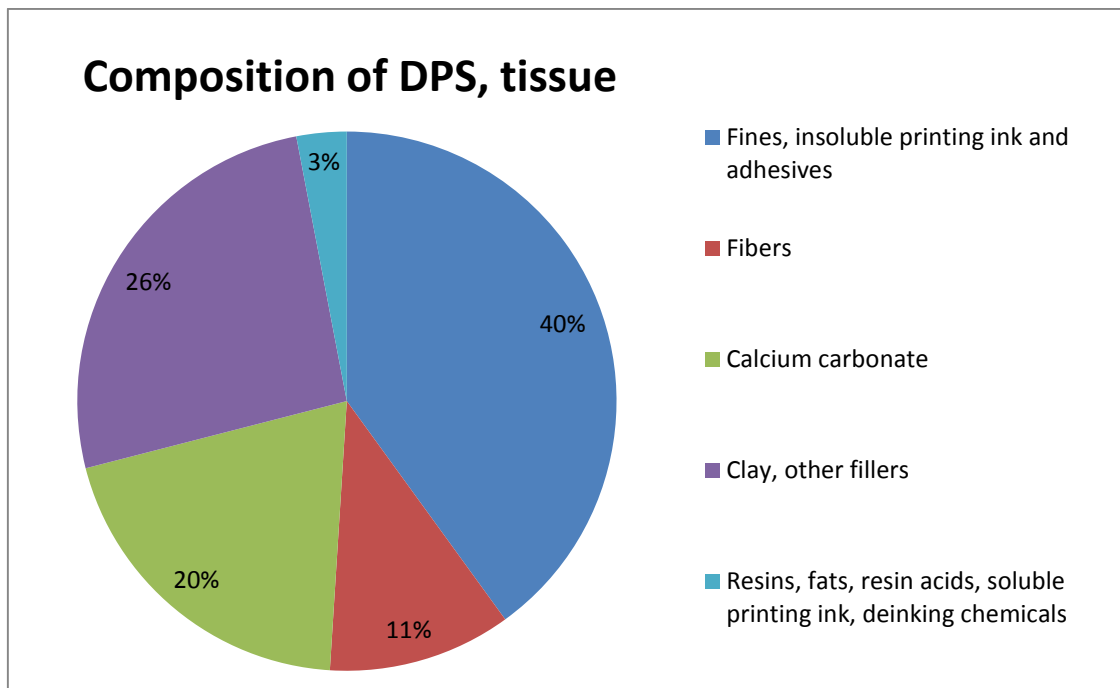
Material and chemical compositions of screening rejects of five German mills producing RCF testliner and corrugated medium showed that solids content of screening reject sludge was 55%. Of the dry solids (DS), fiber represented 36 %, plastics 45% and incombustibles (inorganic matter) 18 %. (Göttsching & Pakarinen eds., 2000, p.512.) Even though this stream still includes a high proportion of plastic contaminants, it also includes relatively high amounts of valuable substances like fiber and fillers.

Deinking sludge is a mixture of fillers and pigments, fibers, fiber fines, printing inks and adhesive components. Its dry content can vary from 38 to 62 %, and the average is 51 %. The ash content of deinking sludge varies from 36 to 67 percent of the dry substance. The fiber proportion is relatively low at 7 % in the RCF intended for graphic paper production. In the production of hygienic paper, fiber loss is bigger resulting in fiber proportion of 11 percent. The compositions of the two different deinking sludges are presented in Graph 1 and Graph 2. (Göttsching & Pakarinen eds., 2000, p. 512-514.)





**Graph 1.** Composition of dry substances in deinking sludge from wood-containing graphic paper production (Göttsching & Pakarinen eds., 2000, p. 513).



**Graph 2.** Composition of dry substances in deinking sludge from hygienic paper production (Göttsching & Pakarinen eds., 2000, p. 513).

From Graph 1 it can be seen that inorganic components like fillers and coating pigments compose 55 % of the dry solids. Roughly, the sludge can be divided into three categories: organic, inorganic and volatile. Organic content includes sections of fines and fibers,

inorganic content referring to calcium carbonate ( $\text{CaCO}_3$  or CC) and clay, volatiles being fats and acids. As seen in Graph 2, in tissue production including wash deinking, the proportion of inorganics is lower at 46 %, due to a higher amount of organics.

The research program by Waste & Resources Action Programme, also referred to as WRAP, studied possibilities of sludge utilization as a value-added ingredient in other industries. The sludge used in the study was collected from Aylesford mill in the U.K., and originated from recycled newsprint production (Rothwell & Éclair-Heath, 2007, p. 60-61). The sludge was treated with the KDS Micronex process, more specifically described in Chapter 4.2.1. The composition of the Aylesford sludge is presented in Table 1, both before and after processing.

**Table 1.** Composition of Aylesford sludge prior and after KDS Micronex processing as bone-dry solids (Rothwell & Éclair-Heath, 2007, p. 60-61).

Component	Proportion (DS)
<b>raw sludge</b>	
<i>Fiber</i>	36 %
<i>Filler</i>	64 %
<b>After Micronex processing</b>	
<i>organic</i>	34 %
<i>calcium carbonate</i>	29 %
<i>china clay</i>	37 %

When comparing the deinking sludge composition in Graph 1 to the Aylesford sludge composition, one may notice that the organic content is quite the same, but proportions of  $\text{CaCO}_3$  and clay are quite the contrary. The difference can be explained by usage of  $\text{CaCO}_3$  growing significantly during the last decade, and use of kaolin being reduced.

Because of the refining level of the raw material, deinking sludge can potentially have higher pollutant levels than rejects from conventional sludge from WWTP. Huge variations in the pollutant content of recovered paper may also contribute to concentrations of common pollutants in de-inked pulp. In Table 2, pollutant contents of deinking sludge and sludge from municipal WWTP are presented. From Table 2, it can be seen that pollutant levels in the deinking sludge are quite the same as in the WWTP sludge, excluding copper and zinc which might have significantly higher concentrations. On the other hand, comparison of deinking sludge to pulp and paper mill WWTP sludge would be more

reasonable than comparing it with municipal WWTP sludge, as the contaminants are significantly different to begin with. For comparison, the average concentrations of these heavy metals found in natural soils are also listed.

**Table 2.** Contents of typical pollutants in deinking, municipal WWTP sludge and soil (IPPC, 2001, p. 251).

<b>Component</b>	<b>Unit</b>	<b>Deinking sludge</b>	<b>WWTP sludge</b>	<b>Average soil concentration</b>
Cadmium (Cd)	<i>mg/kg<sub>DS</sub></i>	0,02 -1,54	< 0,1	0,01-2
Mercury (Hg)	<i>mg/kg<sub>DS</sub></i>	0,1 - 0,9	< 0,1	0,01-5
Copper (Cu)	<i>mg/kg<sub>DS</sub></i>	64 -345	40	15-40
Zinc (Zn)	<i>mg/kg<sub>DS</sub></i>	34 - 1320	250	50-100
Lead (Pb)	<i>mg/kg<sub>DS</sub></i>	9,5 - 79,4	30	15-30
Nickel (Ni)	<i>mg/kg<sub>DS</sub></i>	< 10 - 31	10	15-30
Chromium (Cr)	<i>mg/kg<sub>DS</sub></i>	4,8 - 96,6	10	50-200
Volatile solids	<i>% of DS</i>	33 - 64	48	-

## 2.3 Valuable components of sludge

Recognition of potentially valuable waste streams may be difficult and there is no definitive guidance on the matter. Reduction of waste ending up in a landfill is of course valuable in itself, but distinction e.g. if a stream in focus is more valuable in combustion or in material reuse is hard to establish. When it comes to the deinking process, in the beginning the reject consist mainly of objects, metals and coarse components, as also in wastewater treatment. As explained in Chapter 2.2., the screening rejects includes fibers to some extent but still greater is the proportion of plastics. After mechanical processing, the fiber slurry can be considered clean from external impurities like plastics. Because of the combustible plastics material, screening rejects have a heating value of over 20 GJ/t of DS, whereas deinking sludge provides average heating value of below 7 GJ/t of DS (Göttsching & Pakarinen eds., 2000, p.512). This could be one possible parameter to help draw the line.

### 2.3.1 Fiber and fiber fines

The fibers in recycled paper sludge can be described as lignocellulosic (Krigstin & Sain, 2008, p. 9). The length, area or quality parameters of fibers in DPS haven't been studied a lot. It is known, that e.g. aging of ink increases fiber losses in deinking, but whether this

affects long fibers or fines, is unknown (Göttsching & Pakarinen eds., 2000, p. 400). It is also not always clear how much wood-originated material there is in the sludge and what is the size range of fines.

Some distinction can be made with a screen of 200-mesh which would mean screen opening of about 74  $\mu\text{m}$ . Particles that pass the 200-mesh screen are considered fine. This is supported by Göttsching and Pakarinen (2000, p. 401) stating that the proportion of fiber fraction (+ 200 mesh) in the accept of flotation deinking increases and that removal rate of wood-based fines (-200 mesh) is proportional. This would imply that a fiber fraction of +200 mesh accumulates in the accept flow and that finer material is more likely to end up in the reject. Geng et al. (2007, p. 348) noted that PS contains more long ( $> 54 \mu\text{m}$ ) fibers than DPS and that DPS contains more particles with small area, which supports the previous statement.

Anyhow, it is quite insignificant what the exact size distribution in paper mill sludge is, as the separation of fibers from other components is not reasonable because of technological and economical obstacles. In the end, the value of the fibers and fines in the sludge is their contribution to organic material, some flexibility and duration properties and lightness they can provide to the end product.

### **2.3.2 Fillers and coating pigments**

Calcium carbonate (CC) and kaolin clay are the two mineral constituents most commonly used in paper making. Calcium carbonate ( $\text{CaCO}_3$ ) is used in fillers and coating pigments can be further divided into grounded (GCC) and precipitated (PCC) calcium carbonate (Omya AG, 2011). Also, talc (magnesium silicates) and titanium oxide  $\text{TiO}_2$  can be used. A study by INGEDE showed that calcium carbonate was removed more readily than kaolin in the flotation step, its reduction being two times higher than clay (Göttsching & Pakarinen eds., 2000, p. 401).

Ash of the sludge consists mainly of inert materials, such as clay, titanium dioxide and calcium carbonate. There has been some discussion of filler recovery by a method called wet air oxidation (WAO) which is designed to oxidize the organic matter, leaving the

inorganic fraction unharmed. However, in pilots, problems have occurred with brightness of the recovered fillers. The process has been anyway practiced in some mills in the United States to reduce the sludge volume. (Wiegand & Unwin, 1994, p.92.) In studies that examined deinking sludge ash composition, i.e. the inorganic portion of the sludge, it has been learned to consist mainly of  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{CaO}$  and  $\text{TiO}_2$  oxides (Davis 2003, p. 47).

### **3 UTILIZATION POTENTIAL IN INDUSTRIAL PRODUCTS**

The research on paper mill sludge and deinking residue reuse potential in different types of products has included several composite and construction materials. Some utilization possibilities have been studied by the US Environmental Protection Agency, EPA. In these studies, possibilities of reuse e.g. as an absorbent material in wastewater treatment or as ready-mixed concrete have been explored. (Kay M., 2003, p. 20.) The production processes and raw materials of these applications are presented in this chapter, along with reuse possibilities in theory and in practice.

The industrial reutilization possibilities can be divided into two categories – ones benefiting from high fiber and low inorganic content of sludge, the other ones requiring high inorganic content and a minimum amount of organic compounds. In the first category, there are applications like corrugated material and moulded pulp production, hardboard, asphalt and insulation board. The latter includes building products like brick, cement and building blocks. (Krigstin & Sain, 2008, p.14.)

Some trials have been made between the pulp and paper industry and other industry fields to find out the technical suitability of sludge in the production. For these trials, the sludge was processed with KDS Micronex –cyclone moisture content of 10 to 15 %, and separated to the fiber and filler fractions. These fractions or raw wet sludge was then supplied to potential customers. (Rothwell & Éclair-Heath, 2007.) The results from these trials are discussed in the corresponding chapters. The KDS Micronex equipment is explained in detail in Chapter 4.2.1.

#### **3.1 Fiber-based products**

In this chapter, the industry fields benefiting from organic or fibrous content of the sludge are presented. These include the products of the pulp and paper industry and fiber-based building materials, like fiberboard and millboard.

### 3.1.1 Fiberboard products

Particleboard is a term for a panel manufactured from lignocellulosic, i.e. wood-based, particles or pieces combined with synthetic resin or other binder and bonded under heat and pressure. The term particle in particleboard traditionally refers to chips or flakes, particles larger than individual fibers. In particleboard, the interparticle bond is created by the binder. Boards, which are mainly composed of lignocellulosic fibers, are called fibrous-felted board, respectively. In fibrous-felted products, the integral bonds are created by interfelting fibers. Hardboard is a fibrous-felted board product compressed to high density. The basic difference between hardboard and other boards is that hardboard uses wood lignin as the main binder. Fiberboard means a dry formed panel manufactured from fibers with a synthetic resin. (Maloney, 1993, p. 26.)

In paper mill sludge, and especially in deinking sludge, the amount of intact fibers is small, as discussed in the previous chapter. The sludge, however, has high fines content, the proportion of fines (200 mesh) being 37 % according to Davis et al. (2003, p. 1). The particle size affects the structure, internal bonding and therefore properties of panels produced. In respect to definitions by Maloney (1993), a board produced from sludge with high fines content and a synthetic resin would be called “fiberboard”.

The use of deinking sludge in the production of medium density fiberboard, also known as MDF, has been studied by Geng et al. (2007) and Davis et al. (2003). Fiberboard panel is one of the most popular materials used in furniture and building applications (Geng et al, 2007, p.346). MDF is the most typical fiberboard and is usually used in the furniture industry, wall paneling or e.g. in dashboards and inner doors of cars.

The composition of fiberboard can vary greatly. In a study by Davis et al. (2003), phenol-formaldehyde resin was used, the rest of dry solids was fiber (or partially clay and  $\text{CaCO}_3$  when DPS was used). Geng et al. (2007) used urea-formaldehyde resin with fiber mixtures. A board made from glass and cellulose fibers and synthetic resin could consist of 20 % resin and 80 % fiber of which 25 to 50 % cellulose, and the rest glass fibers (Bullock, 1984). Fiberboard compositions and composition of DPS, if known, used are presented in Table 3.

**Table 3.** Compositions of fiberboards in studies by Davis et al. (2003) and Geng et al. (2007) and the corresponding deinking sludge compositions.

<b>Fiberboard composition</b>	<b>Davis et al.</b>	<b>Geng et al.</b>
<i>Resin</i>	6 %	12 %
<i>Virgin fiber</i>	47 %	70 %
<i>DPS</i>	47 %	30 %
<i>  fines of DPS</i>	35 %	-
<i>  clay + CaCO<sub>3</sub> of DPS</i>	20 + 4%	54 %

Davis et al. (2003, p. 49-50) tested various different furnish compositions, at the highest the CaCO<sub>3</sub> content of the furnish was 4 % of the oven dried weight whereas clay content was 20 %. These proportions were determined regarding the composition of DPS in question. The deinking sludge consisted of 35 % fines, 20 % clay and 4 % CaCO<sub>3</sub>. The ratio of DPS to virgin fiber was 50:50. Davis et al.(2003, p. 47) states that the short fiber content of primary sludge fills gaps between virgin fibers providing hardboard with increased bending strength. It was noticed that increased clay content worsened all of the studied mechanical properties. CaCO<sub>3</sub> or fines content were found to have no significant effect on mechanical properties. The study concluded that clay content should be decreased or eliminated with preliminary treatment of the deinking sludge to improve properties of DPS/MDF board. Increasing resin levels or finding binders better suitable for coating clay were suggested. (Davis et al. 2003, p. 54.)

The calcium carbonate content of deinking sludge has increased in the last ten years, while use of kaolin has reduced. Even though the study showed that CaCO<sub>3</sub> had no significant affect on mechanical properties, the proportion of CaCO<sub>3</sub> was relatively low compared to the clay content and it is likely, that if the CaCO<sub>3</sub> proportion was raised to 20 % it would affect the mechanical properties as well. Most likely the best possibility to enable DPS use in fiberboard production would be the reduction of filler content, but at least it is now known that a 4 % CaCO<sub>3</sub> proportion doesn't affect mechanical properties significantly.

When comparing the physical and mechanical property requirements of MDF (ANSI, 2002, p. 7) to the results by Davis et al. (2003, p. 51-52) one may notice, that in the clay content of 5 % OD weight, ANSI requirements are met. The result attained with a clay content of 20 % doesn't fulfill the requirements.



In the study by Geng et al. (2007), spruce-pine-fir, also referred to as SPF, fiber was mixed with DPS and PS at weight ratios of 3:7 or 7:3 and 12 % resin. Also in this study it was discussed that a high content of inorganic substances in DPS could decrease the strength of fiberboard. A PS/SPF mixture, in ratio of 7:3, fulfilled ANSI requirements of mechanical properties set for MDF grade 120. Also a DPS/SPF mixture, weight ratio 3:7, met the same ANSI requirements. As a conclusion of the study, the PS had more and longer fibers than did DPS and therefore had more suitable fiber structure for fiberboard manufacturing and provided MDF panels with better mechanical properties than DPS, and was considered to have excellent potential in the fiberboard manufacturing process. (Geng X. et al, 2007, p.347-350.)

Even though Geng et al. (2007) presented that PS showed much better potential in MDF manufacturing than did DPS, these results can be considered really promising. The 3:7 ratio of DPS:SPF means that DPS contributed 30 % of the raw materials. Compared to maximum contents of about 5 % e.g. in the ceramics industry this is a big proportion.

Davis (2003) and Geng et al. (2007) both stated that inorganic content, especially clay, reduces mechanical properties of MDF. Otherwise there seemed to be no physical or operational hindrances to prevent DPS use as an addition in fiberboard production. In the latter study a fiberboard incorporating DPS even managed to meet ANSI standards. A small proportion of inorganic or 35 % proportion of fines didn't have significant effect on board properties. This implies that if the filler content were at least partially removed from the sludge, utilization of DPS in MDF production could be a viable opportunity. No allowed concentrations or threshold limits of trace element heavy metals in fiberboard were found in literature.

A company called Homasote produces multipurpose panels for building and sound proofing purposes. The panel is produced of 100 % post-consumer recycled newsprint, with paraffin wax working as the binder. (Ecohaus Inc., 2009.) According to Davis et al. (2003, p. 47) there is a plant in Turkey manufacturing hardboard incorporating primary sludge to be used as core stock in furniture manufacturing.

### 3.1.2 Moulded pulp

Moulded pulp is traditionally manufactured from recycled fiber. The pulp is formed on a mould, water partially removed by vacuum to form the shape after which the product is dried e.g. in an oven. This application requires a maximum ash content of 10 %. (Rothwell & Éclair-Heath, 2007, p. 68.)

West Fraser Timber's Alberta Newsprint Company supplied in 1997 seven percent of its sludge to manufacturing of molding egg cartons according to Glenn (1997, p. 36). No further information of this or more specific characterization of the sludge was however found.

### 3.1.3 Millboard

Millboard is a generic term referring to various high density and thickness board products. It can mean hard board with high density which is manufactured from 100 % recycled fiber. The grammage of millboard varies from 1000 to 5000 g/m<sup>2</sup>. It differs from corrugated board by caliper and contains no cavities, while usually millboard is manufactured by calendering. It has uses e.g. in the automobile industry, shoe industry, furniture and luggage products. This type of use requires high fiber content and low ash content to guarantee wanted caliper. (Rothwell & Éclair-Heath, 2007, p. 69.)

Trials to use sludge as an additive in millboard production were run with the fiber fraction of Aylesford paper mill sludge from recycled newsprint production. The composition of the fiber fraction is presented in Table 4. (Rothwell & Éclair-Heath, 2007.)

The incorporation of fiber fraction into millboard, in order to replace about 5 % of recycled fiber obtained from old corrugated containers, was successful. The final product was found to perform identically to the existing product and no influence on the final product properties were noticed. (Rothwell & Éclair-Heath, 2007, p. 76.)

**Table 4.** Composition and other characteristics of fiber fraction (Rothwell & Éclair-Heath, 2007, p. 62).

<b>Characteristic</b>	<b>Typical specification</b>
<i>Moisture</i>	10 %
<i>Fibrous and organic matter</i>	47 %
<i>Approximal fiber content (of organic matter)</i>	50 %
<i>Filler (clay and CaCO<sub>3</sub>)</i>	53 %
<i>Fiber length</i>	2,0 mm
<i>Gross caloric value</i>	2490 kWh/tonne

Millboard can also refer to bituminized paperboard that is used in auto panels, or to roofing felt which is paperboard impregnated with tar, bitumen or asphalt. Asphalt, or composite, shingle for roofing for example can be produced with organic felt mat, a base made of cellulose fibers of recycled paper formed into sheets. In shingle production, a roll of organic felt is mounted, dried and presaturated, after which it is coated with hot asphalt. (Hall, 2011b.)

The term can also refer to ceramic fiber millboard used in insulation. Ceramic fiber millboard consists of ceramic fibers, clay, insert fillers and a small amount of organic or inorganic binder for better strength. Kaolin, a naturally occurring high-purity alumina-silica fireclay, is suitable for fiber raw material. Ceramic fiber millboard can be used in high-temperature insulation and heating applications. (Intersource USA Inc., 2011; Thermal Ceramics, 2011.) This type of product could make use of the filler content, but should most likely have low or even zero content of organics to be fire proof.

### **3.1.4 Softboard**

Softboard is used in insulation and fireproof applications, as well as in pin boards. The raw material consists ideally of short fibers and has an ash content below 10 % for optimal thickness. Often softboard is manufactured from old newspapers. Softboard is produced from a thick wet pulp blanket which passes through a high intensity press and a drying tunnel. (Rothwell & Éclair-Heath, 2007, p. 68.)

Test trials to use the Aylesford mill fiber fraction, presented in Table 4, in softboard production were conducted in the U.K. The final product was found to be weaker and denser than the existing product. The customer was willing to continue trials, if an ash content of below 10 % was achieved. (Rothwell & Éclair-Heath, 2007, p. 77.)

### **3.2 Mineral-based products**

Reuse possibilities of deinking sludge and other recovered paper residuals depends on the composition of inorganic compounds. Common inorganics in deinking sludge are calcium carbonate and clay. In the combustion ash of deinking sludge, calcium oxide and sintered clay are primary components.

Generally, three methods had been used, in 1994, to use (paper mill) sludge in building material industry. One of these was the sludge use as a feedstock to cement kiln. Another option was to use sludge in cementitious composites, where the use of organic fibers would increase the durability of the product and reduce cracking related to shrinking. In these studies it was concluded that combining Portland cement with deinking sludge could contribute to a composite material suitable for building blocks, wallboards, panels and such. Also use of sludge in the production of lightweight aggregate was studied. (Wiegand & Unwin, 1994.)

#### **3.2.1 Cement and cementitious products**

Cement is a universal term meaning binder, a material that sets independently binding other materials together. There are different kinds of cements, others that are called hydraulic because they harden as a result of hydration, an inorganic chemical reaction which provides e.g. Portland cement its strength. Lime and gypsum plaster can be considered to be non-hydraulic cement, as they develop strength only in dry circumstances.

The basic raw materials in the production of cement are limestone ( $\text{CaCO}_3$ ), clay, sand and iron ore. The proportion of limestone in kiln feed is about 80 to 90 % and clay 10 to 15 % (British geological survey, 2005). According to Achternbosch et al. (2005) proportions of  $\text{CaCO}_3$  and clay in raw meal are about 80 and 20 %. Also silica and aluminum can be

beneficial in the process. When the raw materials are burned at a temperature of 1400 to 1500 °C, they form calcium, silicon, aluminum and iron oxides (Göttsching & Pakarinen eds., 2000, p.538). The hard substance from the kiln is called clinker and is mixed with gypsum to produce Portland cement. The gypsum prevents cement from flash setting. Usually, the proportions are 95% of clinker to 5 % gypsum (British geological survey, 2005).

Residues from wastewater treatment plant (WWTP) that are high in inorganics can contain significant quantities of these substances. Residues from recycled paper manufacturing consist mainly of inorganics, and the deinking sludge can provide components like silicon dioxide and aluminum which are beneficial for the process. Boiler ash from the wood and WWTP residues are found to be suitable for cement and brick manufacture, as long as the carbon content of the ash is below 6 %. In Portland cement production, the fly ash of newsprint mill is used. Ash can contribute to the process as a source of calcium, aluminum and silica. (Bird & Talberth, 2008, p. 12, 19, 22.)

The main ingredients of cement production,  $\text{CaCO}_3$  and clay, are present in deinking sludge in high proportions, so theoretically the sludge could be beneficial material to the process. The problems of reuse are considered mainly economical, caused by the volume of water in the sludge increasing transport costs and the low price of virgin raw materials (Rothwell & Éclair-Heath, 2007, p. 67).

According to Göttsching and Pakarinen (2000) both deinking sludge and ash from combustion of deinking sludge can be used as secondary raw material to the kiln. Deinking sludge ash from fluidized bed combustion can also be used as a hydraulic additive to cement clinker (Göttsching & Pakarinen eds., 2000, p.538-540). According to Wiegand and Unwin (1994) in 1994 there was one mill practicing sludge reuse in cement production full-scale. They sent all their primary sludge and coal boiler ash to a cement manufacturer to fulfill 2% of the kilns total feedstock.(Wiegand & Unwin, 1994, p.92)

Champion International's Hamilton plant in Ohio, U.S., used its sludge and boiler ash in 1997 as filler in cement manufacturing (Glenn, 1997, p.36). The sludge Champion International provides to Portland cement manufacturing is primary sludge from a non-

integrated paper mill, dried with a rotary dryer to 85 % solids content (Hardesty & Beer, 1993, p. 815). Because the wastewaters and therefore sludge originates only from the papermaking process, it is expected to include high amount of inorganics and fibers in good condition. The inorganics content of the sludge was a lot like the one seen in Graph 1, 40 % clay and 19 % limestone. However, the fiber proportion was 30 % compared to 7 % deinking sludge in Graph 1 (Hardesty & Beer, 1993, p. 816.) Most likely, there is not a lot of stickies and adhesives in the sludge, making it easier to dry and handle than sludge from a deinking process.

The use of deinking sludge in an application is dependent on how well the materials in sludge correspond to raw material proportions in the targeted application. As one universal recipe for cement production does not exist, the proportions of ingredients may vary. A chemical composition of one type of cement is presented in Table 5 (Gineys et al., 2010). When comparing chemical compounds of deinking sludge ash, discussed in Chapter 2.3.2, to the chemical composition of cement in Table 5, one can see that inorganic part of sludge provides most of compounds present in cement.

**Table 5.** Chemical composition of cement (Gineys et al., 2010).

<b>Compound</b>	<b>wt-%</b>
<i>CaO</i>	68,07
<i>SiO<sub>2</sub></i>	22,98
<i>Al<sub>2</sub>O<sub>3</sub></i>	4,73
<i>Fe<sub>2</sub>O<sub>3</sub></i>	2,72
<i>MgO</i>	0,8
<i>K<sub>2</sub>O</i>	0,69

There are no guidelines or trace element threshold limits for heavy metals in cement or cementitious products. This is probably because trace elements are mostly bound to cement during the hydration and therefore their leachate is minimal and cement isn't considered to be hazardous to humans or to the environment. There was however a study in which the use of waste materials in cement production and their influences on trace elements in cement were examined (Achterbosch et al. 2005). Achterbosch et al. (2005) concluded that trace element concentrations in cement would increase, but no maximum or threshold

values for them were set. Maximum trace element concentrations in Portland cement at present, by literature, are presented in Table 6.

**Table 6.** Maximum trace element concentrations of Portland cement today (Achterbosch et al. 2005).

<b>Trace element</b>	<b>max. ppm</b>
<i>Cadmium (Cd)</i>	6
<i>Mercury (Hg)</i>	-
<i>Copper (Cu)</i>	98
<i>Zinc (Zn)</i>	679
<i>Lead (Pb)</i>	254
<i>Nickel (Ni)</i>	97
<i>Chromium (Cr)</i>	712

### 3.2.1.1 Cement mortar products

Cement mortar is a paste used to bind blocks together in masonry. Fundamentally, it is a mixture of cement, hydrated lime ( $\text{Ca(OH)}_2$ ), aggregate and water. Studies have been made using both metakaolin produced in deinking sludge incineration and of the direct use of sludge in mortar products. As discussed in the previous chapter, ash is already considered to be a suitable raw material for the cement industry, but use for sludge as such is still limited. Yan et al. (2011) have studied the use of sludge as an additive in cement mortar products and the effects on the physical and mechanical properties of the mortar. Cellulose is a known retarding concrete admixture, and in the study 20 weight-% of sludge in the mixture delayed final setting time. The compressive strength with the sludge was 62% of that of the reference, at 20,5 MPa but was considered to be in range for masonry product usage. The sludge didn't appear to make any difference in long-term cement hydration or hardened paste properties. The sludge proportion increased drying shrinkage and the volume of permeable voids in the final product. Dissolved lignin can work as an air-entraining admixture, providing better workability and consistency and freezing durability to the concrete. A conclusion of the study was that up to 2,5 weight-% deinking sludge didn't affect the physical or mechanical properties of the mortar significantly and incorporating deinking sludge can be a favorable supplementary addition to the product. (Yan et al. 2011.)

The sludge studied by Yan et al. had quite high organic matter content, at 53 % dry solids, with ash representing only 47 % (Yan et al., 2011, p.2086). When compared to Graph 1 and Graph 2, the high organic content would imply the deinking sludge came from tissue production or similar. Probably, a deinking sludge generated in graphics paper repulping or newsprint production with a higher proportion of inorganics, would provide better results in a similar test.

### 3.2.1.2 Concrete

Concrete is a composite material consisting of aggregate, usually sand or gravel, mixed with water and cement. Cement works in the mixture as a binder.

Blawaik and Raut performed a quite similar test with concrete and waste paper pulp than Yan et al. did with mortar and sludge. Their conclusions were that compressive, splitting tensile and flexural strength of concrete at 28 days increased up to a 10 % proportion of waste paper pulp. Further mixing of pulp into the concrete reduced all strengths gradually. (Blawaik & Raut, 2011.) As the sludge by Blawaik and Raut also had a high organics proportion, so the conclusions that are possible to be drawn from Yan et al.'s research apply. The detected increase in strengths could be explained by differences in the fiber proportion. For example long, good condition fibers are supposed to give better strength characteristics than short, worn fibers. On the other hand, Blawaiks research only lasted for 28 days, where Yan et al. tested the strengths still after 90 days.

Sludge use as an additive in cement and cementitious products has been studied and implemented to some extent. The results are promising, while e.g. primary sludge already has a use in the cement industry, as does the ash recovered from sludge incineration. The studies about deinking sludge in cement production are somewhat controversial, as some conclude that deinking sludge hinders the development of mechanical strength in products and others that up to a 10 % sludge proportion sludge can be beneficial to products. All the studies however imply that the use of deinking sludge in cementitious products is preferable. It was also promising, that none of the studies reported sludge fractionation to be obligatory, but that the sludge was used as such. It is also good to remember that there are great differences in sludge compositions and that a sludge with high inorganic content



could provide better strength characteristics in products than the ones in studies by Yan et al. or Blawaik and Raut. This is why executing trials and pilots is crucial to promote sludge utilization in these products.

### 3.2.2 Plasterboard

Gypsum is a natural, rock-like mineral, but it is also produced as by-product of some industrial processes, such as flue-gas desulphurization. In the desulphurization process, gaseous sulfur dioxide,  $\text{SO}_2$ , is absorbed into limestone ( $\text{CaCO}_3$ ) or lime ( $\text{Ca(OH)}_2$ ) slurry. Both reactions form calcium sulfite ( $\text{CaSO}_3$ ). In a reaction called forced oxidation, calcium sulfite can be further oxidized to calcium sulphate and therefore gypsum (Venta 1997). This synthetic gypsum, often referred to as FGD gypsum, has been reported to be used as a raw material in wallboard production for 20 years in the U.S (United States Gypsum Corporation, 2009).

As discussed in Chapter 2.2, deinking sludge can consist of up to 34 %  $\text{CaCO}_3$ . In an ideal case, we would be able to separate the  $\text{CaCO}_3$  from the sludge and purify it from contaminants to harness it for the desulphurization process, after which it could be oxidized to gypsum and used in production of building materials. This would however require efficient separation processes that could preserve  $\text{CaCO}_3$  in a viable state, and make it free of impurities.

Gypsum plaster is a building material, used in a series of applications e.g. plasterboard, fibrous plaster and moulds. Drywall is a construction material consisting of thin panels of gypsum board. The primary component of these products is mineral gypsum, in powder form, and its chemical composition is  $\text{CaSO}_4 \cdot 2 \text{H}_2\text{O}$ . The water molecules in gypsum remain in crystalline form until heated to  $100^\circ\text{C}$ . To produce plaster, the gypsum has to be heated to remove water and then rehydrated, adding excess water and additives to form a slurry. To produce plasterboard, this thick slurry is then laminated between two paper boards. The additives are used to change the density of the plaster or to bond the plaster with cardboard. The board is then dried by evaporation or heating at a temperature of up to  $250^\circ\text{C}$ , so that the plaster solidifies. (Miller & Greig, 2008.)

Plaster itself is a brittle substance. Fibrous plaster is a mixture of plaster and fibers, usually glass fibers, to give better strength properties. This is usually moulded and used in slabs. Gypsum plaster is usually used as a fire retardant, which when heated releases water, preventing the fire from spreading. Additives commonly used in plaster products are: starch, grounded gypsum, lignosulphonates, potassium sulphate and detergents. (Miller & Greig, 2008.)

Paper sludge could possibly be utilized as an additive in the plasterboard. In theory, the fibrous material in sludge could improve some strength properties or provide a porous structure to the final product and therefore lower density. According to Hall (2011a), paper pulp is currently added to plaster to improve the tensile strength of the core, yet the composition and type of sludge is not specified. 50 % of gypsum panel is air, to minimize the product weight and to make it easier to work with. The porous structure is produced by foaming agents. Vermiculite or clay may also be added to the product to enhance fire-resistance. (Hall 2011a.)

The fiber proportion could be useful at strength improvement but is possibly harmful for the fire-resistance of the plaster. When produced at high enough temperatures, the organic matter burns leaving pores in the structure. However, most likely the drying temperature of 250 °C isn't high enough. Clay in the sludge could also be used in board production, but should be as pure as possible from contaminants. Hall (2011a) states that each additive of plaster can be a maximum of 0,5 % gypsum powder, so the plasterboard production can, even in theory, provide reuse for only a limited amount of sludge.

Another gypsum product to make with waste paper is gypsum fiberboard, in which there are no conventional board sheets around the gypsum board, but 18 % of the gypsum material is replaced by ground recycled fiber to provide reinforcement (Venta, 1997). Recycled fiber reinforcement of gypsum composites have also been studied by Carvalho et al. (2007) but as in this study the cellulose fiber content came from repulped kraft paper cement bags and can be considered to have quite different composition to deinking sludge. The study can only provide indicative information on mechanical properties of fiber-cellulose composite.

Already in 1996, there was a patent in the United States describing the drywall and building block manufacturing process with deinking by-product reinforcement. The method added deinking by-products at a consistency of 3 to 10 weight-% to stucco. The process was claimed to achieve a deinking-based fiber loading in drywall to be up to 30 weight-%, and to provide better bending strength to the final product. The composition of the by-products was not exclusive explained. (Tran, 1996.)

Rothwell and Éclair-Heath (2007) ran trials with the filler fraction of Aylesford mill sludge to be used in acoustic plasterboard production. The sludge composition and its use in plasterboard are presented in Table 7. The filler content of stucco-filler mixture was at first 10 %, but later the filler content of 5 to 7 % was found to be optimal. Also in these trials it was noted that extra material worked as retardant, the setting time of mixed plasterboard being 11 minutes compared to normal time of about 8 minutes. The results showed that incorporation of filler in plasterboard was successful. It was however speculated whether a purer filler fraction would give even better results. (Rothwell and Éclair-Heath, 2007, p. 78.)

**Table 7.** Composition of acoustic plasterboard incorporating wet Aylesford sludge and sludge composition.

<b>Component</b>	<b>Proportion</b>
<i>Stucco</i>	93-95 %
<i>Wet sludge</i>	5 - 7 %
<i>filler</i>	57,5 %
<i>fiber</i>	31,7 %
<i>water</i>	10,8 %

### **3.2.3 Bricks and ceramic tiles**

The main raw materials of ceramic products are natural clay minerals, such as kaolin, shale and loam. Sand, grog manganese, barium among others can be used as additives to provide shades or other characteristics. Papermaking by-products, sawdust, ammonium compounds, wetting agents and flocculents are also known to be used in brick manufacturing. The processes vary greatly from one another, which is why there is no universal recipe of raw ingredients. The colour of brick is determined by the mineral content. Iron provides brick a red-brown shade, Calcium carbonate makes brick yellowish. Simply put, the manufacturing process consists of grinding the raw material and mixing it

with water, extrusion, drying, firing and cooling. (Rothwell & Éclair-Heath, 2007, p. 66.) In brick production, fiber-containing waste is used to improve porosity and therefore heat insulating properties for the end product. Aside from conventional paper mill sludge, deinking sludge also has a use as a porosity agent. Sludge as a porosity agent can be replaced by sawdust or pelletized polystyrene. In raw brick material, grained clay and  $\text{CaCO}_3$  can promote rheology. The  $\text{CaCO}_3$  proportion in raw brick material can be 30 volume-% at the highest. To produce bricks with equivalent compressive strength, up to 20 volume-% of sludge, 10 volume-% polystyrene and 5 volume-% of sawmill dust can be used as porosity causing material. (Göttsching & Pakarinen eds., 2000, p.541-542.)

Both calcium carbonate and clay provide advantages to brick manufacturing, e.g.  $\text{CaCO}_3$  decreases fluorine and sulfur emissions during burning. A too high amount of porosity causing and biologically active sludge on the other hand are harmful in brick manufacturing. (Göttsching & Pakarinen eds., 2000, p.541-542.) This implies that deinking sludge with a high inorganic and lower organic content could be more beneficial to the process than conventional sludge. On the other hand the fibrous content of sludge reinforces bricks and significantly lowers the risk of tearing in the drying process (Göttsching & Pakarinen eds., 2000, p.541).

Weng et al. (2002) have studied brick manufacturing from the dried sludge of an industrial wastewater treatment plant. They concluded that up to 20 % sludge content can be added to bricks as a clay replacement, 10 % sludge content being ideal and that the brick fired at a temperature of 880 to 960 °C met the requirements of Chinese National Standards (Weng et al., 2002.) Quite interestingly, the origins of the dried sludge used in this research were not specified.

Rothwell and Éclair-Heath (2007) reported that untreated, wet Aylesford sludge was supplied to two brick manufacturers in the U.K. One customer asked for deinking sludge in particular, without wastewater treatment effluents as biological activity can cause odors and be harmful to the brick manufacturing process. This customer added the sludge at 2 – 3 % addition rate to the bricks to replace the fuel ash content and assessed that Aylesford sludge can be used as an ash substitute up to 7 % of total wet brick weight. It was also estimated in laboratory trials ran by this customer, that a sludge having similar  $\text{CaCO}_3$

content could be used as an additive up to 9 % without any affect on the final brick shade. Another customer used one percent sludge in the overall brick weight. Both concluded that sludge didn't appear to change the appearance or physical properties of the bricks. (Rothwell and Éclair-Heath, 2007, p. 79-80.) Especially valuable information from these trials is that even though the customers were first offered the filler fraction, they both ended up using the raw wet sludge.

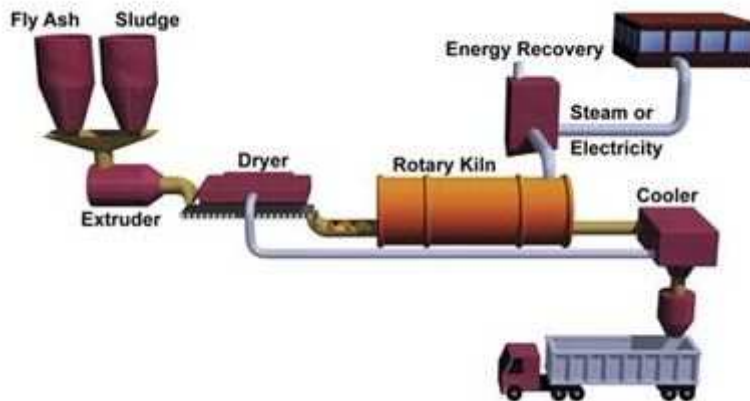
Similar to bricks, ceramic tiles can be produced from kaolin, silica, feldspar, clay, wollastonite and chalk. These products would most likely require high purity and cleanliness of minerals, which is why it is more often referred to as a possible utilization target of coating kitchen wastes collected as a separate waste stream without discharge to the wastewater treatment plant. (Valtonen et al., 2000, p. 24.)

There are also companies called DuraStone and Congoleum, which are manufacturing tiles and flooring from limestone composites. Tiles are composed of organic binder which contains vinyl resins, plasticizer additives. Ten percent or more of vinyl content is recycled. Limestone represents 85 % of the tile. (GreenFloors, 2005.)

### **3.2.4 Lightweight and glass aggregate**

In the building industry, aggregate is a term describing fillers of construction materials. Aggregates are used in cementitious products like concrete, masonry, building blocks and asphalt. In concrete production, sand is an example of an aggregate material. Lightweight aggregates, also known as LWA, serve as a density-reducer in the final product without affecting the strength properties. In 1994, one pilot had used a proprietary mixture of paper mill sludge, ash and additives to produce LWA and was planning on building a large-scale pilot. (Wiegand & Unwin, 1994, p. 92-93.)

Between 1994 and 2000 Minergy Corporation produced LWA from fly ash of a coal combustion plant, paper mill sludge from a coated paper mill and some municipal WW sludge and sold it for construction and concrete producers. Production ended when the properties of the ash changed due to new fuel so that it is now directly used in the concrete industry. Minergy's LWA production process is presented in figure 3.



**Figure 3.** Minergy Corporates LWA production process (Minergy Corporation Limited, 2011).

Liaw et al. (1998) have studied paper sludge use in lightweight aggregates, and noticed that cement products using these aggregates have similar or even superior physical properties compared to commercial aggregates. Some mechanical properties, like compressive strength, however decreased. (Liaw et al., 1998.)

The mineral material in flotation sludge is also used in the glass aggregate production. The organic fraction is combusted and the inorganic material separated by vitrification. Minergy Corporation has developed a technology to transform sludge into material which can be used in sandblasting grit, abrasives, roofing shingle granules and as a cement additive. Trace metals are immobilized into the product. In production, modified cyclone furnaces are used. The Minergy glass aggregate facility operates in Wisconsin, U.S. and processes 1 300 tons of sludge per day. (Minergy Corporation Limited, 2011.)

### 3.2.5 Rockwool

Rockwool, also known as mineral wool, is produced from a mixture of coke, diabase and limestone melted at a temperature of about 1600 °C and lead to spinning wheels, where the material forms mineral fibers from 1 to 10 μm diameter. Other igneous rocks like basalt and dolomite may also be used. Some phenol-formaldehyde glue and mineral oils are added to the process. Phenol-formaldehyde resin works as a binder, as in fiberboard production, to glue the fibers together but leaving air gaps in between. The volume of voids in the mineral wool can vary from 95 to 99 % of the total volume. (Valtonen et al., 2000; Berge, 2009.)

Rockwool International was taking part in the EU Life -program in 2008 and had a campaign to promote waste recycling into raw ingredients of rockwool production. The required origins or compositions of waste were undetermined, but Rockwool production was advertised to utilize 400 000 tons of waste per year, e.g. from the aluminum industry. The goal was to substitute 25 % natural rock raw material with residue materials from other industries. Both inorganic and combustible waste, such as ash, sand and metals were suggested as potential raw materials. The organic waste would contribute to the process as energy, and inorganics with the right chemical composition as a substitute for the virgin stone material. (Rockwool Group, 2008.)

### **3.3 Other products**

There are various products mentioned in research and reports considering sludge reuse. Many of these can't be divided into products which would benefit from the fibrous or inorganic components of sludge. These and other miscellaneous products will be discussed in this chapter.

#### **3.3.1 Fiber cement**

Fiber cement is a composite material made of sand, cement and cellulose fibers. Previously the fibers used were asbestos fibers, but as the negative health impacts of asbestos were discovered this led to the prohibition of asbestos in building products. Asbestos was then replaced mainly by wood fibers. In Finland, the use of asbestos was prohibited in 1994 (University of Helsinki, 2011). Fiber cement is used in cladding and exterior siding as well as tile underlay on decks and bathrooms. Fibers provide the product with a higher resistance to cracking and a degree of flexibility as well as lighter weight.

The inorganic raw materials of fiber cement can include Portland cement, silica, silica fume, limestone, metakaolin, fly ash and calcium silicate. The organic components may include kraft cellulose pulp, recycled fibers, synthetic fibers, flocculants and defoamers. (Moslemi, 2008, p. 117.)

After restrictions on asbestos use, various fibers including chemical and mechanical wood pulp, polypropylene, glass and mineral fibers were considered. Fibers from chemical wood pulping were found to comply best with requirements which were e.g. ability to withstand alkalinity, temperature resistance and fiber mechanical properties. The only factor against the use of chemically pulped fiber was its price. (Moslemi, 2008.) Recycled fiber is a cheaper ingredient than virgin fibers, so they might be a solution to this problem.

Moslemi (2008) also studied the impact of fiber length on the fracture toughness of the resulting fiber cement. It was noticed that the higher the fiber content of over 0.74 mm long fibers, the higher the fracture toughness of the product. Fibers with length of 0.33 mm didn't improve toughness significantly even at high proportions. (Moslemi, 2008, p. 116.) This is why knowing the fiber length distribution of deinking sludge is important, as findings by Moslemi are most likely true in other fiber-based applications too. As seen in Table 4, the average fiber length of Aylesford sludge was found to be 2.00 mm, which would, according to study by Moslemi give significantly better toughness properties for the final product. These results would imply that a sludge equivalent to Aylesford sludge could be beneficial in fiber cement production. However, Moslemi (2008) also investigated the properties of virgin and two different types of recycled fibers and their size distribution and concluded that over 95 % of recycled fiber is below 3.00 mm in length. For virgin fiber this proportion is about 45 % (Moslemi, 2008, p. 116).

### **3.3.2 Thermoplastic and rubber composites**

Polypropylene is a thermoplastic polymer. It is used e.g. in the production of carpets, plastics, containers and automotive components. In a study by Girones et al. (2010) a paper mill sludge with 30 % fibers and 70 % CaCO<sub>3</sub>, dried to water content below 15 % was used as a filler and reinforcement for polypropylene composite. In plastic processing, mineral fillers and fibers (e.g. fiberglass) are used to improve the mechanical properties and economical efficiency. Polypropylene was mixed with sludge at 25, 37.5 and 50 weight-% based on dry weight. The mixture was granulated, homogenized, injected and then conditioned after which tensile, flexural and impact tests were carried out.



The results showed that the tensile strength of composites diminished when the proportion of sludge grew, as was to be expected by studies that investigated the addition of fillers to thermoplastics. There has been discussion of fiber, hydrophilic by nature, failing to provide effective reinforcement to the hydrophobic polypropylene matrix. This is why polypropylene based coupling agents have been used to form covalent bonds between polypropylene and natural fibers. Addition of a coupling agent (at a rate of 4 weight-%) was noticed to prevent a decrease in tensile strength almost completely. Flexural strength increased with sludge content, even without the coupling agent. The study concluded that a composite formulation including 50 % sludge and coupling agent provided similar tensile and increased flexural strength compared to plain polypropylene and can be used as a substitute for mineral-filled composites in applications of low mechanical requirements. (Girones et al., 2010.) Theoretically, deinking sludge could be beneficial for the process if the hydrophilic nature of fibers was noticed to be a problem, while most of the matter in deinking sludge is hydrophobic and therefore removed in the flotation process at the first place.

Ludvik et al. (2007) studied the influences of cellulose fiber and bentonite clay in the production of biodegradable thermoplastic composites. In this study, the biodegradable thermoplastics tested were polylactic acid, also known as PLA and polybutelene adipate, PBAT. The cellulose fibers were dispersed into an aqueous gel of sodium bentonite clay, which was then combined with powdered PLA and calcium carbonate filler. Then the composite was dried, extruded and injection molded to make thin parts. The study concluded that composites had good moisture resistance and higher modulus than plain thermoplastics. Some challenges occurred with the fiber-thermoplastic matrix, but it was discussed that these could be overcome with surface modification. (Ludvik et al., 2007.)

As discussed previously, Girones et al. (2010) overcame the fiber-plastic bonding issues with coupling agents. In mechanical strength tests of thermoplastic composites by Ludvik et al. (2007, p. 255) it was seen that longer fibers provided better strength characteristics than short fibers. These results support the statements by Moslemi (2008) considering fiber cement, as discussed in chapter 3.3.1. Theoretically, the deinking sludge can provide raw material to all of these composites, but it must be considered if the reinforcement of the

sludge mixture with some proportion of virgin fiber could provide enhanced strength characteristics to final products.

Also Ismail et al. (2008) have studied paper sludge use in natural rubber composites. In this study, paper sludge was combined with commercial filler, such as carbon black or silica to produce so called hybrid filler which could provide better matrix properties and mechanical properties of final products than the sludge alone. Partial replacement of paper sludge with carbon black was found to improve some mechanical properties, while carbon black interacts with natural rubber. (Ismail et al. 2008.) In this study, the origins of paper sludge were not mentioned. But if carbon black can provide better interaction characteristics to fiber-rubber matrix, then deinking sludge should be beneficial to this process while carbon black is often used in inks as a colorant and therefore present in deinking sludge.

Yet another possible application, which could benefit from calcium carbonate content of sludge is sheet moulding compounds, also known as SMC, which consist mainly of glass fibers, calcium carbonate as filler and unsaturated polyester resins. (DSM, 2011.)

### **3.3.3 Board and loose insulation products**

The different types of board insulation are glass fiber, expanded polystyrene, extruded polystyrene, phenolic foam, polyurethane and polyisocyanurate. As the raw materials of insulation boards and such building products are mostly polymerous by nature, or consist of mineral based products, like glass fiber, direct reuse of sludge is unlikely. This is largely due to the organic content of sludge, which is detrimental to the desirable characteristics of an insulating product, such as fire resistance, conductivity and resistance to absorb moisture. Nevertheless, paper mill sludge could be used indirectly by adding it to the raw materials of insulation boards, like to glass fiber production or polymer composites discussed in chapters 3.2.4 and 3.3.2.

Rothwell and Éclair-Heath (2007) offered the filler fraction of Aylesford sludge to a customer manufacturing insulation sheets. The customer ran tests with the material and

found that the proportion of organics were 31,2 %, which exceeded their acceptable organic content levels, so the trial was suspended.

### **3.3.4 Paving and fibrous road surfacing additives**

Recycled fiber can be used in bitumen modifiers to achieve better properties for road surfacing materials. The fiber additive is known to reduce noise, and provide enhanced binding, elasticity and durability to the pavements. Rothwell and Éclair-Heath provided the fiber fraction of their research to a fibrous road surfacing manufacturer. The customer ran laboratory trials and found that the product failed the bitumen drainage test, and that for their products, a material with a moisture content below 7 % with an ash content below 20 % was required. (Rothwell & Éclair-Heath, 2007, p. 68, 76.)

Another study about sludge use as an additive in stone mastic asphalt mixtures was done by Mari et al. (2009). Sludge contents were set from 0,2 to 0,5 %. It was concluded that with a sludge content of 0,3 to 0,5 % and asphalt content of 5 to 6 %, all mixtures from 4 different paper mill sludges passed the Philippine road pavement specifications. However, the best-performing sludge was provided to the additive at a dry solids content of almost 94 % with an organic content of 66,5 % (Mari et al, 2009, p. 31), which differs quite a lot from composition of deinking sludge. There was nonetheless also a sludge with an inorganic content of over 60 % that likewise passed the requirements. (Mari et al., 2009.)

The additive use of sludge can be considered as one of the applications that should be tested differently for the specific sludge type available. In these applications, the drying properties of sludge will most likely be the limiting factor for sludge reuse.

### **3.3.5 Absorbent material and animal bedding**

WWTP residuals have been used as raw material in industrial absorbents which are available in market (Bird & Talberth, 2008, p. 18). For example, in Washington, U.S., a company called International Absorbent Inc. has used paper mill sludges in oil absorbent materials since the 90's (Glenn, 1997, p. 36).

In 1996, a Wisconsin company called Thermo Fibergen had developed a system to recover long fibers and extract water from the sludge to use the fibers in papermaking and after that the material of short fibers and fillers were to be used as oil and grease absorbents, cat litter and granules. They had bought a company called GranTek which already had a continuous operation to take sludge at a consistency of 40 percent solids, dry it and form granules to be further dried and screened. (Glenn, 1997, p. 36.) As sludge from a recycled paper process already includes relatively few long fibers, it might be a suitable raw material for these type of products, if it could be easily dried.

Marcal Paper in New Jersey, U.S., established a series of absorbent products, called Kaofin, for industrial floor cleaning and oil and grease spill prevention and non-industrial purposes manufactured from their recycled paper waste sludge in the 90's (Glenn, 1997, p.36). In 1994 they had a conflict with the New Jersey Department of Environmental Protection (NJDEP) whether Kaofin is a recycled product or it should be considered a solid waste. The Marcal Paper managed to get an exemption to Kaofin from solid waste and were granted permission to market their product with some limitations of quantity of material in the end use product. High purity criteria and control of the product quality and safety were set for the product. In the late 90's Kaofin had another law suit about unsuitable use as landfill covers and was determined as solid waste to be disposed because of violations on previous exemption. (Justia, 2008.) It seems that the argument between Marcal Paper and NJDEP still remains unsolved. No Kaofin products are marketed or sold nowadays.

WWTP residuals have been used as raw material in animal bedding products which are available in the market (Bird & Talberth, 2008, p. 18). International Absorbent Inc. has also produced animal bedding products from sludge. The precise production process is not explained on their internet site, but products are claimed to be produced from reclaimed cellulose fiber, either short fiber cellulose pulp or pre-consumer long cellulose fibers. Some cat litter products are clay based. (International Absorbents Inc., 2005)

### **3.3.6 Filler reuse in papermaking after oxidation**

Two emerging technologies have been mentioned in discussions about recovering the filler content of the paper mill waste sludge by burning all organic substance of sludge. The first one is the wet air oxidation process discussed in chapter 2.3.2, and the other one supercritical water oxidation, SCWO. A few example, for which full-scale trial data is available, use thermal or wet air oxidation to destroy the organic fragment to recover inorganic fillers as ash (Wiegand & Unwin, 1994, p.92). There have been promising results for this technology with sludge from a Swedish mill. The filler brightness achieved was as good as 74% ISO and paper properties were similar between the papers with virgin and recovered filler. (Jernström, 2005, p. 14.)

## 4 POSSIBILITIES TO ENHANCE REUSE POTENTIAL

There are several reasons why there hasn't been large scale reuse of paper mill effluents so far. Aside from the technical challenges, economical reasons such as low price of virgin materials or costs of running required pilots and lack of standards, specifications and actual demand for recycling, have hindered the development of recycling sludge as a construction material (Kay M., 2003, p.20).

As discussed in Chapter 3, many of the utilization possibilities could benefit from a higher purity of the sludge components. For example, when incorporating the fiber fraction of the study by Rothwell and Éclair-Heath (2007), three of four trials with potential customers concluded that a higher fiber content and lower ash content were crucial in their production.

### 4.1 Current limitations of utilization

Some potential customers of Rothwell and Éclair-Heath (2007) stated that the Aylesford sludge incorporated components that were harmful for their products or product quality. Many customers were willing to make further trials if their requirements were met. In Table 8, some applications, for which paper mill sludge was found to be unsuitable, are listed. The list is combined from trials or studies that concluded sludge reuse to be possible, if unwanted impurities or components could be eliminated. Respectively, the limiting factors of sludge reuse and desired purity levels are presented in Table 8.

**Table 8.** Limiting factors and minimum requirements set to sludge composition for utilization of sludge in some industry applications.

<b>Application</b>	<b>Limiting factor</b>	<b>Requirements</b>	<b>Reference</b>
<i>Moulded pulp</i>	ash content	< 10 % ash	Rothwell & Éclair-Heath, 2007
<i>MDF</i>	clay content	<< 20 % clay	Davis et al., 2003
<i>Softboard</i>	ash content	< 10 % ash	Rothwell & Éclair-Heath, 2007
<i>Cement mortar</i>	cellulose/ organic content	-	Yan et al. 2011
<i>Plasterboard</i>	organic content	> 65 % filler	Rothwell & Éclair-Heath, 2007
<i>Insulation board</i>	organic content	-	Rothwell & Éclair-Heath, 2007

From Table 8 it can be seen that the limiting factors of sludge reuse are related to the composition of sludge. These have been discussed in detail in Chapter 3, in corresponding products. The applications that could use the fiber proportion of sludge would generally require the ash content of the sludge to be below 10 %. In some studies, it was discussed whether a higher inorganic proportion could provide better product quality, yet no limits for organic content were set.

According to Rothwell and Éclair-Heath (2007, p. 77) one customer reported that sludge had an unacceptably bad smell. Quite surprisingly, the nature of the sludge wasn't mentioned to be a problem in any other of the studies relating to this research. Problems with the characteristics of sludge could have been expected as the deinking sludge is sticky by nature and embodies a relatively high amount of adhesives, chemicals and such substances. However, in general, the physical characteristics of sludge didn't seem to cause much harm.

When it comes to hygienic requirements or presence of trace element heavy metals in sludge, there was not much of discussion about the subject in the research reports. This is probably because there currently is no wide use of sludge in consumer materials. The hygienic requirements would most likely apply to moulded pulp or board products used in packaging, if larger scale sludge reuse was ever implemented. The production of ceramics and most of the building products usually involves drying or burning at high temperatures, so the hygienic quality of the product isn't supposed to be that unsteady. The heavy metals in deinking sludge, discussed in Chapter 2, could become a discussion topic if sludge was implemented on a large scale in e.g. building products. At present, there seems to be no guidelines or limiting threshold values for trace elements in building boards, concrete or such. And even if there were limits, the amounts of sludge to be mixed with virgin ingredients is nowadays so marginal, they most likely would not be exceeded.

## **4.2 Technological aspects and possibilities**

In this chapter, a few promising technologies, that aim to separate and recover valuable components from deinking sludge, are introduced. First is the KDS Micronex, a cyclone which is claimed to produce a dry, fluffy substance from sludge that can be further

separated into separate fiber and filler fractions. The second is the ECO pigment recovery process, designed to recover filler from effluent treatment sludge producing sludge with over 80 % filler content. Other installations designed for recovering components from different types of sludge, are the FilRec system by Ahlstrom and the RECClaim process by ECC International.

Recovering valuable components from the mixed sludge is a process often referred to as fractionation. By today, more studies have been made to fractionate components from one another without affecting the quality of other components. These studies have used separation techniques such as hydro cyclones, centrifuges, belt filtration or sieve bands (Kyllönen et al. 2010, p. 19).

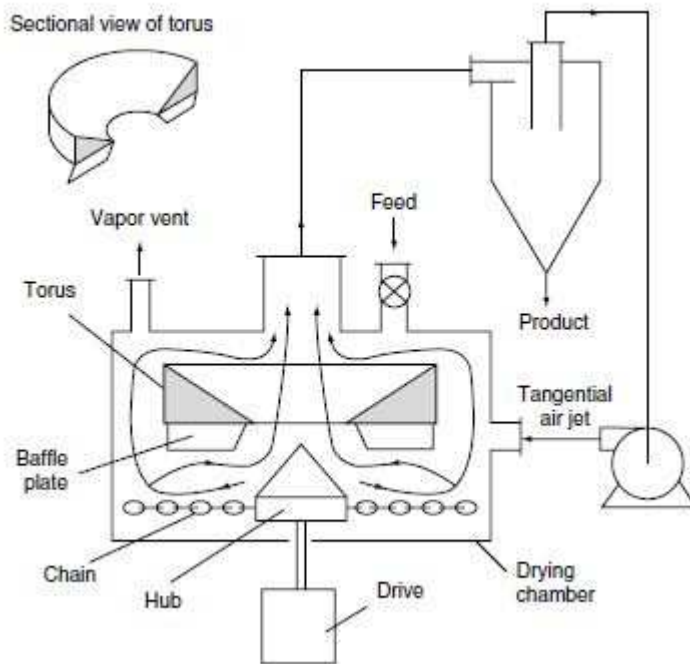
Another important objective of sludge treatment is dewatering. Dewatering aims to reduce the sludge volume. If some of the previously introduced reutilization possibilities were implemented on a large scale, and sludge was transported between the pulp mills and the end user, minimizing the sludge volume would play an important role in economical efficiency. Additionally, in trials to incorporate sludge into plasterboard and polypropylene composites, the moisture content of the sludge was below 15 %. (Rothwell & Éclair-Heath, 2007; Girones et al., 2010).

#### **4.2.1 The KDS Micronex system**

Trials with the KDS Micronex system have showed that a moisture content of a deinking sludge, which is originally about 38 %, can be reduced to 16 % (First American Scientific Corp., 2009). According to Mujumbar (2007) an initial moisture content of 53 % wet basis can be reduced to 22 %. Micronex is classified as a grinder-dryer process. Micronex doesn't have a heat input, the drying happens mainly as a side-effect of the grinding. The chamber of the Micronex encloses 8 spinning chains connected to a hub. The tip speed of the chains is 200 m/s. The feed material is led to the hub after which the centrifugal acceleration flings the material to the chamber walls. Centrifugal and Coriolis forces combined with collisions against the walls and baffle plates cause the material to be pulled apart and broken up. Because of these impacts, the particles are heated simultaneously with increased evaporating properties of matter, leading to an escape of moisture from material.



The moisture is immediately recondensed to fine mist, which is discharged through a vapor vent. The airflow gets blown out of the chamber, carrying particles with it, and led to a cyclone. The conventional cyclone then separates particles from the air, discharging dry, fluffy product at the cyclone bottom. (Mujumbar, 2007.) The schematic diagram of Micronex system is presented in Figure 4.



**Figure 4.** Schematic diagram of KDS Micronex system (Mujumbar, 2007).

The KDS Micronex system itself doesn't include fractionation of the sludge components, so it has to be done as a separate process step. However, First American Scientific Corporation (2009) claims that further fractionation is not possible with conventional drying methods. Rothwell and Éclair-Heath (2007) used the KDS Micronex combined with vibrating screen separation to produce separate fiber and filler fractions from the sludge. The fiber fraction was presented previously in Table 4 and the composition of filler fraction produced is presented in Table 9.

**Table 9.** Composition and other characteristics of filler fraction (Rothwell & Éclair-Heath, 2007, p. 63).

Characteristic	Typical specification
Moisture	10 %
China clay	30% (of DS)

<i>Calcium carbonate</i>	40% (of DS)
<i>Fiber and organic matter</i>	30% (of DS)
<i>Mean particle size</i>	< 50 micron

The goal of KDS Micronex –processing was to achieve a 75 % purity level, that means below 25 % fiber in the filler fraction and below 25 % inorganics in the fiber fraction. As seen in Table 4 and Table 9, the goal was not reached. The purity of the filler fraction was however quite impressive with only 30 % organic matter. The fiber fraction had only about 24 % actual fibers, and 53 % of fillers in it. According to Rothwell and Éclair-Heath (2007), the vibrating screen separation process could be further optimized or other screening options could be tried, to improve the fraction purity.

A study by Krigstin and Sain (2008, p. 13) characterized the sludge from three types of paper recycling mills. One produced only newsprint, another tissue and third combined newsprint and tissue. The final solids contents of sludges were 91 to 97 %. Fines were separated from fibers with a 2-mm holed screen. Fines were further sieved with a 40-mesh screen to fractions “fines 40+” and “fines 40-”. Examination of the fractions showed that the inorganic content in the “40-“ fraction was higher than in the KDS effluent before screening. The original sludge sample processed with KDS Micronex, had 34 % inorganic content. Both of the “fines 40-“ fractions had an inorganic content of about 40%. The fiber fractions total inorganic content was 20,9 % and “fines 40+” had an inorganic content of 22 to 27 %. The reduction of calcium carbonate in fiber fraction was found to be successful, as the original amount of 16,6 % was reduced to 8,5 %. As a conclusion, Krigstin and Sain found that the sludge prepared by KDS Micronex had potential to produce a high purity fraction when further dry fractionation methods were used. (Krigstin & Sain, 2008, p.13-14.)

Both of the studies confirm that inorganic material tends to accumulate in the fines fraction. However, the purity of both fractions wasn't that good in any of the studies. Rothwell and Éclair-Heath succeeded to produce a filler fraction with 70 % inorganic content from the sludge with an original inorganic content of 64 %. Krigstin and Sain managed to increase the organic content from the original 66 % to about 79 %. It can however be assumed that with an optimized screening technology, from raw sludge with high inorganic content, a quite pure inorganic fraction or from high organic content sludge, a satisfyingly pure organic fraction, respectively, could be refined. Another fraction would

most likely have poor purity and was therefore unsuitable for reuse. It has been proposed by First American Scientific Corporation (2009) that after Micronex processing and filler separation, the residual fiber fraction was used as fuel in an incineration process.

Both of the studies also confirmed that impressively low moisture contents were achieved. Krigstin and Sain managed to get a solids content of 97 % at the highest. Moisture contents of fiber and filler fractions of Aylesford sludge were 6 and 3 percent at the lowest, respectively. Rothwell and Éclair-Heath (2007, p. 45) proved also that the drier sludge, the higher the purity of the fiber fraction.

#### **4.2.2 The ECO pigment recovery system**

The ECO plant in Oulu, Finland, is a process to recover pigment from the primary clarifier effluent sludge of a paper mill. The recovered pigment, referred to as ECO pigment can then be used as filler in multicoated fine paper production. (Jortama, 2003.) Even though the process was not developed for deinking sludge, it was a novelty process in 2003 and produced high purity pigment fraction, which was successfully used as filler in paper production, so the utilization attempts of deinking sludge could benefit from the knowledge in the study by Jortama (2003).

The ECO process consists of sludge disintegration, separation of fillers from fibers and other impurities with a wire washer, fractionation, chemical treatment of the recovered material and a separate handling process for combustible rejects. Disintegration of sludge is done by a blade wheel. The wire washer is operated contrary to conventional sludge thickening, as the pigment passing the wire is the accept flow and the fibers and other solids stuck on wire are the rejects. The wire washer used was called OptiThick GapWashed, manufactured by Metso. The fractionation step consists of vibrating screens and centrifugal cleaners. The vibrating screens are, however, normally bypassed when producing the ECO filler for paper machines. The centrifugal cleaning was done with a two stage process including LC cleaners by Andritz. The cleaning is supposed to remove difficult, small dirt specks e.g. resins, sand and inks. Chemical treatment is made primarily with peracetic acid, providing an ECO pigment of up to 88 % ISO brightness. (Jortama, 2003, p. 63-72.)

The ECO system managed to accumulate particles below size 45  $\mu\text{m}$  in the accept quite well, the proportion of these fines being about 96 %. The ash content of the wire washer accept was high, mainly around 85 %, although it was over 70 % already in the original sludge. The inorganic proportion consisted of 84 %  $\text{CaCO}_3$ , 8 % clay, 6 % aluminum oxide and 2 % talc. The dry solids content of ECO pigment was usually between 2 to 6 %. (Jortama 2003, p. 82-96.)

The pigment recovery process by Jortama (2003) had quite different objectives than did the research made with KDS Micronex. Both can however be beneficial in future sludge separation technology. The KDS Micronex tries to dry the sludge first and fractionate it afterwards, while the ECO process is designed to fractionate sludge at low consistency. Both have their advantages and disadvantages, the low consistency ECO process providing high purity filler fraction that could most likely be quite easily dried afterwards. Implementing the ECO process for deinking sludge would probably be difficult because of stickies and other impurities adhering fillers to the fibers. The product from KDS Micronex however is dry and components can be separated to some extent. The dry screen and sieve separation processes are not yet efficient enough to provide high purity fiber and filler fraction, as stated in previous chapter. Especially the grinding feature of Micronex is quite harmful for sludge quality, while retaining long, intact fibers would be beneficial in both separation processes and utilization of sludge as fiber reinforcement.

### **4.3 Economical aspects**

Some successful trials were made with the KDS Micronex processed sludge in the study by Rothwell and Éclair-Heath (2007). As discussed previously, transporting and utilization of the sludge is usually more economic, the lower the moisture content of the sludge. Assuming that the dried sludge from the KDS Micronex processing should be utilized, some rough calculations could be done to discuss the profitability of the operation.

As there is no obvious return from investment for KDS Micronex, the investment cost of the equipment should be calculated with an annuity method. For the calculations, a few assumptions were made which are presented in Table 10. The investment cost of Micronex

was 250 000 £ in 2007, and sludge input to process 30 000 t/a (Rothwell and Éclair-Heath, 2007, p. 89). An investment cost of 290 000€ for KDS Micronex was calculated at a currency exchange rate of 1,16 Euros to a Pound (GBP). Electricity consumption for the Micronex system is 100 kWh/ton of input sludge (First American Scientific Corp., 2009). The average industrial electricity price, in 2011, in Finland was 0,076 €/kWh and the equivalent price in e.g. Germany would be 0,125 €/kWh (European Commission, 2011). The in-service life and interest rate were approximated. It was assumed that no extra personnel was needed to operate the machine, and that there were no extra expenses for maintenance.

**Table 10.** Assumptions made for investment calculations of sludge drying with KDS Micronex.

Parameter	Unit	Value
<i>Investment cost</i>	N [€]	290 000
<i>Interest rate</i>	r [%]	8
<i>In-service life</i>	T [a]	5
<i>Sludge input</i>	$q_m$ [t/a]	30 000
<i>Electricity price</i>	[€/kWh]	0,075
<i>Electricity consumption</i>	[kWh/t <sub>input</sub> ]	100

Using the annuity method, machine cost for each year of operation can be calculated from Equation 1.

$$A = N \cdot \frac{r \cdot (1+r)^T}{(1+r)^T - 1} = 290\,000\text{€} \cdot \frac{0,08 \cdot (1+0,08)^5}{(1+0,08)^5 - 1} = 72\,632\text{€/a} \quad (1)$$

Sludge treatment cost per unit of sludge treated with Micronex can be further calculated from Equation 2. If the in-service life was expected to be 3 years, the unit cost of input sludge would be 3,75 €/t.

$$\frac{A}{q_m} = \frac{72\,630\text{€/a}}{30\,000\text{ t/a}} = 2,42\text{ €/t} \quad (2)$$

Energy costs per unit of sludge processed can then be estimated from Equation 3.

$$100 \frac{\text{kWh}}{\text{t}} \cdot 0,076 \frac{\text{€}}{\text{kWh}} = 7,6\text{ €/t} \quad (3)$$

With the electricity price of Germany, which is closer to the European average, the sludge drying costs of operation would be 12,5 € per ton of input sludge. When the unit cost of the machine investment is added, the treatment cost per ton of input sludge in Finland and Germany would be about 10 €/t and 15 €/t, respectively. After reduction of moisture content from 36 % to 15 %, 1 ton of input sludge is equivalent to 753 kg of output sludge. Accordingly, unit costs per 1 ton of sludge produced are 13.3 €/t and 19.8 €/t in Finland and in Germany. Considering the assumptions made, the real costs including e.g. maintenance and operating personnel salaries etc., could be estimated to be about 30 to 40 € per ton sludge produced. For the KDS Micronex investment to be profitable, this amount of money must be profited when selling the sludge to an end user, or saved in landfilling fees.

The price of one ton of bulk calcium carbonate can be estimated to be 180 £, or about 210 € (Pitt, 2002). Assuming that there won't be much additional costs to operating the Micronex and that transportation could be arranged economically, the sludge could be sold at 50 € per ton. This would also require that calcium carbonate and other raw materials haven't gotten much cheaper since 2002, and that sludge would actually benefit the end user somehow so they would be willing to pay for it.

As a comparison, the costs of landfilling in Finland and in Germany are about 50 to 60 € and 30 to 50 € per ton of sludge, respectively (Hogg, 2001). These can be expected to be somewhat cheaper for industry customers, but the fees have most likely risen in the last ten years, so the price for a industrial customer today can be estimated to be about the same that the cost for private customer ten years ago. Therefore the landfill fee for one ton of sludge could be estimated to be somewhat equivalent to the estimated sludge processing cost. This would mean that all the sludge processed with Micronex should be then disposed elsewhere than to a landfill for Micronex processing to be feasible. On the other hand, processed sludge has smaller moisture content and therefore volume, which means saving in landfilling fees. It must also be taken into account that for example in Finland, in 2001, altogether 50 000 tons of deinking sludge was disposed. So most likely there is no single mill with 30 000 tons of annual sludge generation. A large recycled pulp mill in central

Europe then again could probably provide that amount of sludge for Micronex processing and therefore reutilization.

As a proposal for further discussion, the investment on KDS Micronex, or equivalent machine, can be considered for primary sludge drying equipment by providing high solid content sludge with lower transportation costs. This could be done on basis of one or two certain customers. The existing drying system would then provide the sludge producer the possibility to further fractionate the sludge and then try it with customers that have higher quality standards. Possibly new utilization ways could present themselves after sludge fractions were optimized to maximum purity, which would improve profit potential.

## 5 RESULTS

In this chapter, viable and potential utilization possibilities will be briefly discussed. These include successful trials with deinking or similar sludge rejects. Potential for these utilization methods to solve the waste problem and their implementation on a large scale will also be considered.

The concept of paper mill sludge is quite new and can have various meanings, from residues of the deinking process to a mixture of all types of by-products of the pulp and paper industry including wastewater treatment effluents. Sometimes it refers merely to wastewater treatment by-products, as Evanylo et al. (1999) suggest. Although there was an aim to focus on deinking sludge, it was not always clear what term “paper mill sludge” actually meant in all studies. This is good to remember when interpreting the results.

Deinking sludge is a residual of recycled paper processing and is a mixture of all sludge like rejects generated in various process steps of RCF processing. Deinking sludge is generated in the first flotation process to remove printing inks, stickies, fillers and coating pigments from repulped fiber slurry and in all process steps after flotation. Rejects of preceding steps of flotation tend to tally with municipal waste, and are treated separately from deinking sludge. Generation of waste in recycled fiber processing can be found in Figure 2.

Deinking sludge consists of an inorganic proportion of fillers and coating pigment, commonly calcium carbonate and clay. The inorganic proportion is typically around 50 to 60 % of dry solids. The proportion of fibers is typically about 10 %, and the rest consists of fine organic particles. The moisture content of sludge is typically around 40 %.

Surprisingly, the utilization of deinking sludge as material in other industries and products has been studied widely. Sludge has been used as such or mixed with other paper mill sludge effluents. The applications studied were mostly composite materials in which the sludge could replace virgin materials partially. In a study by Rothwell & Éclair-Heath (2007), sludge was processed with a grinder-drier and fractionated. Trials with potential customers were then done with fractions achieved.



Successful trials with paper mill and deinking sludge are presented in Table 11. The same list with more specific details is presented in Appendix 1. As seen in Table 11 and Appendix 1, in ten industrial applications, paper mill sludge was incorporated successfully. In eight of these studies, the sludge was solely deinking sludge or combined sludge from recycled fiber processing. Two studies with medium density fiberboard and Portland cement incorporated only wastewater treatment residues. These were included in the table for the comparison (MDF) or because the composition of the sludge was considered to be equivalent to that of deinking sludge (Portland cement).

**Table 11.** List of successful trials with paper mill effluent sludges discussed in this report.

<b>Application</b>	<b>Ash content</b>	<b>Organic content</b>
<i>MDF</i>	up to 5 % clay and/or CC	35 % fines
<i>MDF (80 % PS, 20 % SS)</i>	19,5 % of OD weight	-
<i>MDF (DPS)</i>	53,6 % of OD weight	-
<i>Millboard</i>	53 % of DS	47 % of DS
<i>Portland cement</i>	40 % CC, 19 % clay	30 %
<i>Cement mortar</i>	47 % of DS	53 % of DS
<i>Plasterboard</i>	64 % of DS	36 % of DS
<i>Brick</i>	64 % of DS	36 % of DS
<i>Glass aggregate production</i>	-	-
<i>Polypropylene composite</i>	70 % CC of DS	30 % of DS

In chapter 3, utilization possibilities were divided to three groups. The first one was fiber-based products. In this chapter, possibilities of deinking sludge to serve as fiber replacement were discussed. The results showed that deinking sludge has somewhat poor in fibrous content for this purpose. Three of four trials by Rothwell & Éclair-Heath (2007) concluded that the inorganic content of the sludge was too high. Two other studies had however incorporated deinking sludge in fiberboard successfully. Sludge was also successfully incorporated in millboard at a consistency of 5 %.

It was noticed in both fiberboard trials that the mineral content of sludge had negative effects on the boards mechanical properties, but despite this the boards produced fulfilled ANSI requirements. In another study, deinking sludge was actually incorporated at a quite high proportion, replacing almost 30 % of the virgin raw material. This can be considered to be a really promising result, while it has the potential to provide utilization for relatively large amount of sludge. It is important to remember, that the initial cost of setting up

treatment processes to enhance the reuse potential is expensive, and most likely not profitable, if there is only utilization prospects for 10 % of the paper mills waste sludge.

The second product group, mineral-based products, also provided some promising results. For example in cement production, sludge has been used successfully since the 90's. Unfortunately, no cement producers were found that advertised the use of deinking sludge as a raw material, so no data in actual figures was found. It was however learned that usually sludge is accompanied with boiler ash for cement manufacturing process. Rothwell and Éclair-Heath (2007) stated that the obstacle of sludge reuse in cement production was, above all, the transportation costs of sludge with high moisture content. This problem could be solved with a cheap, efficient drying process for sludge and finding a cement manufacturer operating nearby to where the sludge was generated. Hardesty and Beer (1993) had a primary sludge with similar inorganic-organic proportion as deinking sludge that was used in cement production at a solids content of 85 %. Most likely, deinking sludge could serve as well, if only it could be economically dried to the same solids content.

Studies with other cementitious products concluded that up to 2,5 weight-%, sludge had no significant effect on mechanical properties and was a preferable additive to production. The production amount should anyhow be quite big to provide use for a reasonable amount of sludge. This applies also to brick manufacturing, which successfully used sludge to replace 1-7 % of raw material. It was however discussed in other studies, that a sludge content of up to 20 volume-% was possible.

KDS Micronex processed Aylesford sludge by Rothwell and Éclair-Heath (2007) was used successfully in plasterboard production, contributing 5 to 7 % of the raw materials of acoustic plasterboard. However, as discussed in Chapter 4.3, processing of sludge with KDS Micronex is quite expensive and for the process to be cost-effective, there should be a viable utilization application for a great proportion of the sludge generated. This would require one large plasterboard producer or many other small end users nearby the pulp mill.

The last, but not least, applications to utilize sludge were composite materials. These are somewhat newer products, like fiber cement and plastic composites. There were no trials to add sludge into fiber cement yet, but sludge could provide many of the raw materials needed. There was however a drawback with fiber length quality in the recovered fiber pulp and it was discussed whether deinking sludge fibers could provide the reinforcement needed. Surprisingly, the incorporation of sludge into thermoplastic composites had given really positive results, with sludge replacing up to 50 % of the composite without significant loss of strength properties and was recommended for applications with low mechanical requirements.

A few non-conventional sludge utilization and disposal methods were also discovered in this research. One is an existing process by Minergy Corporation, which combusts the flotation sludge in a special furnace to produce a glass aggregate. The process is capable of using 1 300 tons of sludge per day. Oxidation process technologies approach the problem from the point of view of burning the organic content to recover the inorganic one.

The limitations of reuse are technological or economical. For example the sludge has a high moisture content which makes it costly to transport and hard to utilize. In studies finding beneficial end use for sludge, the moisture content was often reduced to about 15 %. This level of moisture content can be achieved e.g. by the KDS Micronex system. Calculations showed that unit costs per ton dry sludge produced would be about 30 to 40 € per ton of sludge. This amount of money should be gained from the customer or saved in landfill fees, for the treatment to be profitable. To conclude, on the basis of the price of  $\text{CaCO}_3$  and average landfill fees, if there was an industrial producer nearby the sludge origin that could incorporate a relatively high proportion of sludge produced, the utilization could be viable.

## **6 CONCLUSIONS AND RECOMMENDATIONS**

The objective of this study was to examine sludge-like wastes from recycled fiber processing and then find utilization possibilities for these sludges as a raw material in other industries. Many studies were found and discussed, in which deinking sludge or sludge with a similar composition to deinking sludge had been used successfully in various building and composite materials. Mainly these would offer utilization for quite a marginal amount of sludge, which affects the profit potential of setting up treatment for sludge. However, the unit costs of sludge treatment, with a machine designed for deinking sludge precisely, were estimated and evaluated to be lower than the virgin raw materials or about the same as the cost of landfilling. It was then concluded that sludge utilization could be a viable option, if customers were found relatively nearby to the sludge origin.

Even though individual studies have proved that deinking or paper mill sludge is a viable reinforcement, additive or composite in various applications, all sludges vary in composition and therefore their suitability to raw material must always be tested with real life trials. In this study, it was targeted to collect results from separate studies in to one report and provide ideas and proposals for further discussion. The sludge producing companies are however the only party, that have the possibility to initiate these trials and thereby enhance sludge utilization as a raw material. To enable trials, co-operation between industry fields must be established. Companies should take initiative and actively find nearby operating producers that use raw materials present in the sludge. Successful large scale trials between industrial operatives would be a significant step towards improved material efficiency.

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APPENDIX 1. Successful trials with paper mill effluent sludge.

<b>Application</b>	<b>Ash content</b>	<b>Organic content</b>	<b>Details</b>	<b>Reference</b>
<i>MDF</i>	up to 5 % clay and/or CC	35 % fines	DPS - virgin fiber ratio 50:50	Davis et al., 2003
<i>MDF (80 % PS, 20 % SS)</i>	19,5 % of OD weight	-	PS:SPF ratio 7:3; ANSI A208.2-2002, grade 120	Geng et al., 2007
<i>MDF (DPS)</i>	53,6 % of OD weight	-	DPS:SPF ratio 3:7; ANSI A208.2-2002, grade 120	Geng et al., 2007
<i>Millboard</i>	53 % of DS	47 % of DS	Replaced 5 % of recycled fiber successfully	Rothwell & Éclair-Heath, 2007
<i>Portland cement</i>	40 % CC, 19 % clay	30 %	Primary sludge	Hardesty & Beer, 1993
<i>Cement mortar</i>	47 % of DS	53 % of DS	2,5 weight-% of sludge content	Yan et al. 2011
<i>Plasterboard</i>	64 % of DS	36 % of DS	5 - 7 % dried sludge in stucco, moisture content 10,8 %	Rothwell & Éclair-Heath, 2007
<i>Brick</i>	64 % of DS	36 % of DS	up to 7 % sludge of total brick wet weight	Rothwell & Éclair-Heath, 2007
<i>Glass aggregate production</i>	-	-	1 300 tons of sludge per day	Minergy Corporation Limited, 2011
<i>Polypropylene composite</i>	70 % CC of DS	30 % of DS	50 % sludge content, moisture content below 15 %	Girones et al., 2010