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# Recent Advances in Paper Mill Sludge Management

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## 1. Introduction

The management of wastes, in particular of industrial waste, in an economically and environmentally acceptable manner is one of the most critical issues facing modern industry, mainly due to the increased difficulties in properly locating disposal works and complying with even more stringent environmental quality requirements imposed by legislation. In addition, in recent years the need to achieve sustainable strategies has become of greater concern, also because some traditional disposal options, such as landfill, are progressively restricted, and in some cases banned, by legislation. Therefore, the development of innovative systems to maximize recovery of useful materials and/or energy in a sustainable way has become necessary. Industrial wastes are generated through different industrial processes or energy production utilities as additional materials. Industrial symbiosis theory defines non-deliberately produced material as by-products or valuable raw materials which can be exploited in other industrial avenues. Paper industry is a strategic industry in many countries but in the same time, the production of paper consumes high quantities of energy, chemicals and wood pulp. Consequently, the paper production industry produces high environmental emission levels mainly as CO<sub>2</sub> due to energy consumption, or solid waste streams which include wastewater treatment sludges, lime mud, lime slaker grits, green liquor dregs, boiler and furnace ash, scrubber sludges and wood processing residuals. In terms of volume, most solids or liquids are those from the treatment of effluents, although waste from wood is also produced in large quantities.

In this chapter the different processes and technologies for conversion of paper mill sludge (PMS) into valuable products with the special emphasis on the technology for conversion of PMS into absorbent for oil spills sanitation, are discussed. The environmental impact of the latest is evaluated with Life Cycle Assessment (LCA).

## 2. Trends in paper mill sludge generation

The pulp and paper industry produce over 304 million tons of paper per year. In 2005, solely in Europe, 99.3 million tons of paper was produced which generated 11 million tons of waste and represented 11 % residue in relationship to the total paper production. The production of recycled paper, during the same period, was 47.3 million tons, generating 7.7 million tons of solid waste which represented 16 % of the total production from this raw

material (Monte et al., 2009). Global production in the pulp and paper industry is expected to increase by 77 % by 2020 and over 66 % of paper will be recycled at the same time (Lacour, 2005). On average, the majority of waste generated from paper production and recycling is PMS, which is a by-product of up to 23.4 % per a unit of produced paper, the quantity depending on paper production process (Miner, 1991). The countries joined in the CEPI organization itself produce more than 4.7 million tons of PMS per year and global production of PMS was predicted to rise over the next 50 years by between 48 and 86 % above the current level (Mabee and Roy, 2003). This represents an enormous environmental burden due as more than 69 % of the generated PMS is landfill disposed (Progress in Paper Recycling Staff, 1993; Mabee and Roy, 2003). Flowchart of PMS generation is shown on Figure 1.

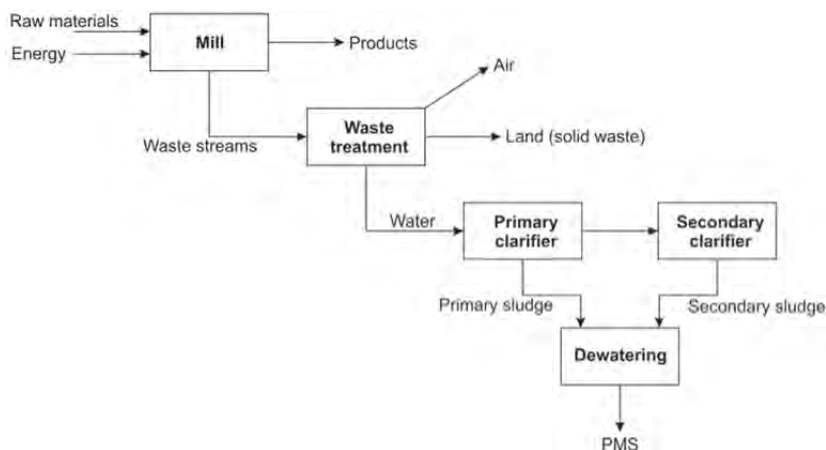


Fig. 1. Scheme of PMS generation

	PMS origin	Organic content wt %	Ash content wt %	Heat value MJ/kg	CEC cmol/kg
Low-ash sludges	Primary sludge from pulp mill producing pulp from virgin wood	94,31	5,69	20,1	30,20
	Primary sludge from paper mill producing paper from virgin wood	93,79	6,21	19,8	32,41
High-ash sludges	Secondary sludge from paper mill producing paper fom recycled cellulose without deinking process	67,23	32,77	16,5	17,30
	Primary sludge from paper mill producing paper from recycled cellulose without deinking process	64,72	35,28	14,2	33,58
	Deinking sludge from papermill that recycled paper not recycled previously	60,36	39,64	12	18,12
	Deinking sludge from papermill producing newspaper	59,30	40,70	12,2	19,06

Table 1. Composition, heating values and cation exchange characteristics (CEC) for different PMS origins (Méndez et al.; 2009).

Recyclers are producing two to four times more sludge as Virgin pulp mills but the characteristics of PMS vary, depending on pulp and paper mill processes and from raw materials entering into those processes. Produced sludge can be considered to fall into two main types: high-ash sludge (> 30% dry weight) and low-ash sludge (< 30% dry weight). High-ash sludges are chemical flocculation sludges generated by pulp mills, primary sludges generated by production of paper from recycled fibers and deinking sludges generated by paper mills, alternatively, low-ash sludge represents primary, secondary or biological sludges generated by pulp or paper mills (Table 1).

Primary and deinking PMS constitutes a mixture of short cellulosic fibers and inorganic fillers, such as calcium carbonate, china clay, and residual chemicals dissolved in the water (Figure 2).

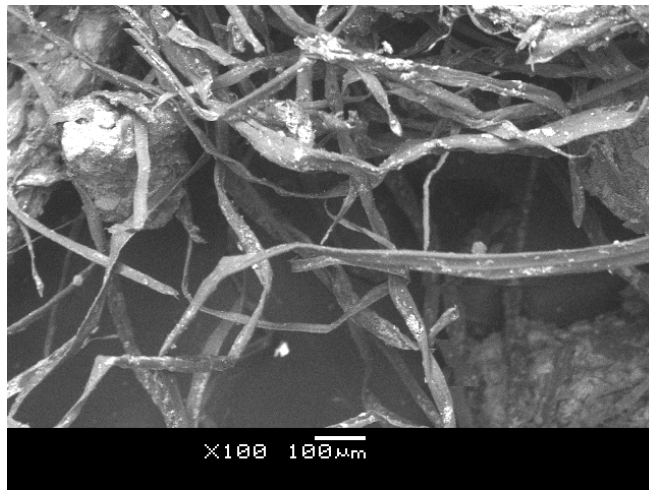


Fig. 2. SEM image show complex structure of PMS (cellulose fibers and china clay).

Currently, PMS are considered to be a waste material. Increased recycling over the past two decades has consequently increased the amount of material that requires disposal. The paper industry implements several methods to dispose of the sludge that pulp and paper production generates (Figure 3).

### 3. Major concerns with paper mill sludge management

#### 3.1 Landfilling

PMS with high organic content (Table 1) in landfill is subjected to aerobic and anaerobic decay. According to Buswell and Mueller (1952), 1 ton of low-ash PMS in landfill theoretically releases into environment approximately 2.69 tons of  $\text{CO}_2$  and 0.24 ton of  $\text{CH}_4$  (Likon et al., 2009). Currently, most PMS is dried, spread or deposited onto the landfill (Mabee, 2001). The landfills can be industrial, in that are constructed and operated by the mills, or they can be independently owned, requiring the mills to pay a "tipping fee" for sludge disposal. The European Landfill Directive (1999/31/EC) and upcoming bio-waste

directive aims to prevent or reduce, as far as possible, the negative effects of waste landfill on the environment, by introducing stringent technical requirements for wastes and landfill. By increasing landfill fees up to 3.5 €/ton annually until the final landfill use fee will reach 40 €/ton. That fact and adaptation of paper mills to Integrated Pollution Prevention and Control Directive 1996/61/CE forced the paper mill industry to look for processes for their waste minimization as well new technologies for waste reuse or environmental friendly disposal.

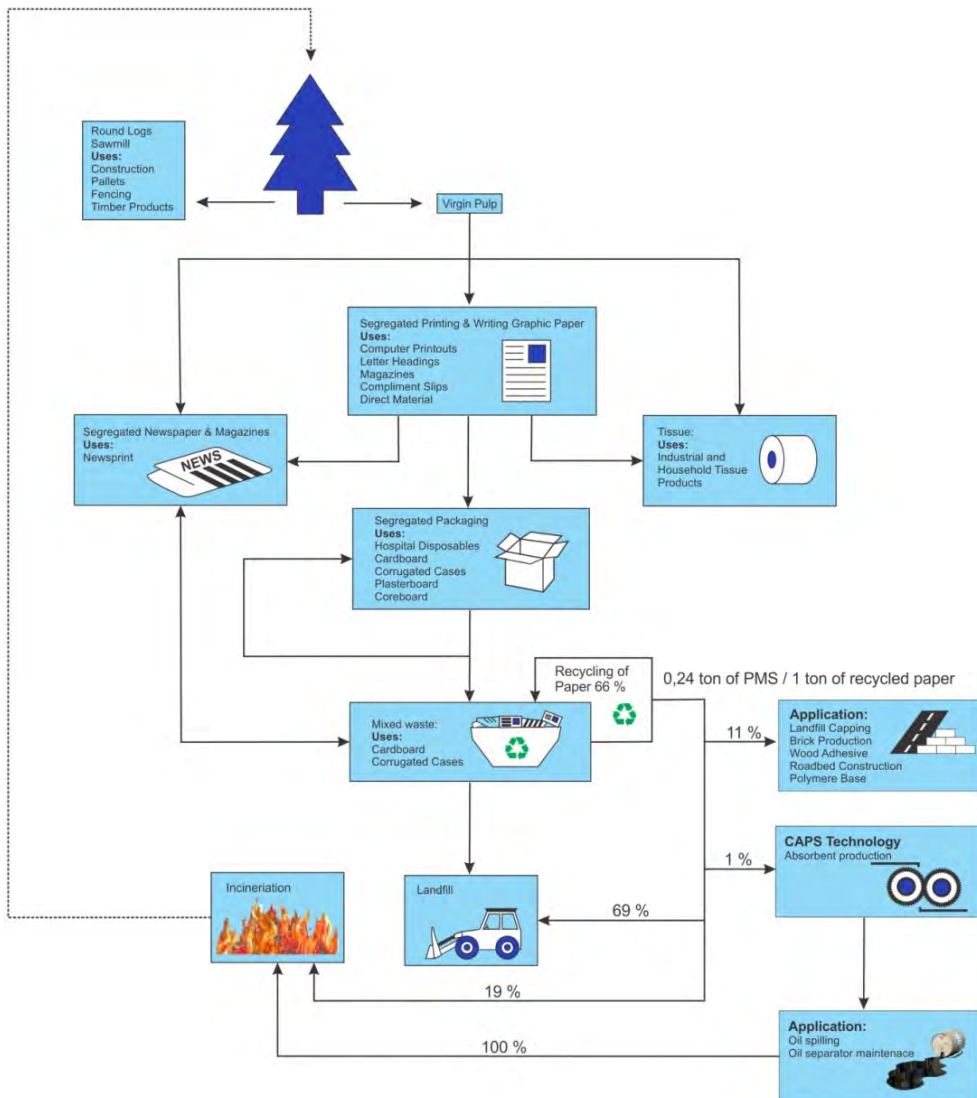


Fig. 3. PMS disposal pathways.

### 3.2 Landspreading

Land surface spreading is one of the possible methods for the application of sludge to forest or agricultural lands for soil enhancement (Christmas, 2002; Ribiero et al., 2010). PMS from pulp mills and secondary sludges are not hazardous due to heavy metals. On the other hand, sludge from deinking and waste paper mills may be relatively high in heavy metals due to the formulations used in ink removal. Compounds that can be found in mill sludges at concentrations above 10 mg/kg of dry sludge include naphthalene, phthalates, chloroform, PCBs, wood extractives or derivatives and chlorinated lignin derivatives (Thacker, 1985). Sludge as land spreading material must be used very carefully due to an unfavorable C:N ratio, high ion exchange capacity and possible Cr toxicity (Norris and Titshall, 2011). With maximum application rates of 12 tons per 405 ha, even a moderately sized mill will need a large area to spread its sludge (Shields et al., 1985; Legendre et al., 2004). The use of land spreading material adjacent to residential areas is questionable due to odor.

### 3.3 Composting

Composting is one of stabilization techniques used for prevention of uncontrolled PMS decay. The use of compost is questionable due to the same reasons mentioned in land spreading. The C:N ratio in the sludge is up to 930:1 and it is not appropriate for plant growth (Thacker, 1985; Scott and Smith, 1995; Monte et al., 2009). Composting of PMS requires a large land area and additional costs for storage and processing (Gea et al., 2005).

### 3.4 PMS for energy

In practice approx. 19 % of PMS is incinerated on the sites due to energy recovery, but the economics of incineration is questionable because PMS contains 30 to 50 % water and only 30 to 50 % of cellulosic fibers calculated on dry solids. For each additional 1 % of moisture content in PMS, the temperature of combustion must be 10°C higher due to process efficiency (Kraft, 1993). The European Waste Incineration Directive 2000/76/EC intends to prevent or reduce, as much as possible, air, water and soil pollution caused by the incineration or co-incineration of waste, reducing at the same time the risk to human health that incineration processes entail. However, under controlled conditions PMS can become a sustainable fuel for co-generation (Figure 4).

Many technologies to utilize different PMS for energy exploitation have been tested and used. That processes include efficient incineration in fluidized bed and circulating fluid bed combustion chambers (Nickull, 1991; Kraft, 1993; Busbin, 1995; Fitzpatrick and Seiler, 1995; Porteous, 2005; Oral et al., 2005) as well as more sophisticated processes for energy recovery as pyrolysis (Frederik et al., 1996; Kay, 2002; Fytili and Zabaniotou, 2008; Lou et al., 2011; Jiang and Ma, 2011), indirect steam gasification (Durai-Swamy et al., 1991; Demirbas, 2007), wet oxidation (Johnson, 1996; Kay, 2002; Maugans and Ellis, 2004; Fytiliand and Zabaniotou, 2008), super critical water oxidation (Modell et al., 1992; Dahlin, 2002; Kay, 2002) and gasification (CANMET, 2005). Advances and drawbacks of energy recovery from PMS are well described in peer reviewed literature (Monte et al., 2009).

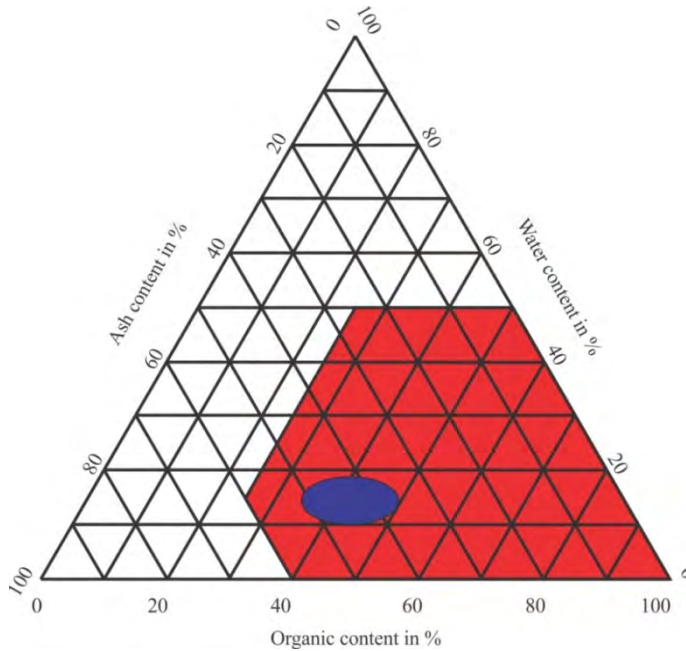


Fig. 4. Fuel triangle for waste from paper industry (blue spot – deinking PMS).

### 3.5 Utilization in brick, light aggregates and cement production

PMS containing a high inorganic fraction (Table 1 – high ash sludges) can be utilized in the production of building materials (Černeck et al., 2005). Due to its combustion matrix, it can be used in the brick production industry. The addition of 5-15 % of PMS as raw materials improves both the final product and the processes. First, since its fiber content increases the porosity of the matrix, it enables the manufacture of lighter bricks; second, it saves fuel in the oven, decreasing firing time and makes the product more resistant against cracking during the drying and firing stages (Monte et al., 2009; Furlani et al., 2011). The same advantages can be used in the production of light aggregates for the building industry. (Ducman and Mirtic, 2011). A similar exploitation has been noticed in cement industry. PMS with high organic content (Table 1 – low ash sludges) has an energy level that makes it an efficient alternative fuel in the production of Portland cement. Currently it is classified as Class 2 (liquid alternative fuels) in the Cembureau classification of alternative fuels (Dunster, 2009). Including up to 20 % of deinking sludge into mortar improves the mortar mechanical properties (Yan et al., 2011).

### 3.6 Landfill capping material

Significant progress has been made in the use of paper mill sludge as a material for land fill cover by replacing the clays or geo-composites. PMS behaves similar to a highly organic soil and has good chemical, hydrodynamic and geotechnical properties which make it an efficient impermeable hydrodynamic barrier for the land field cover (Zule et al., 2007).

Combining the paper mill sludge as hydrodynamic impermeable barrier with metal scoria as an oxidizing layer an efficient landfill capping system can be build. More than 21 different studies have been carried out in 2009 on the use of PMS in the landfill body (Rokainen et al. 2009). The characteristics of PMS have been exploited also for road bed construction for light loaded roads and tennis courts (Moo-Young et al., 1996).

#### **4. Advanced research and utilisation of PMS**

According to modern industrial trends on eco symbiosis economic efficiency is key factor for planning modern technological processes. This includes decreasing the waste streams during production and the use of produced waste as by-products or raw materials with higher added values. As can be seen from Figure 2 PMS is a chemically and physically complex material which can be used in different industrial applications. One of the promising innovations is the use of PMS as heat insulation material with a thermal conductivity factor lower than  $0.055 \text{ W/m}^2\text{K}$ . This is comparable to insulation materials available on the market currently. The US company, Minergy, has implemented technology for incineration of PMS under controlled conditions. The product of incineration is a highly efficient water absorbent composed of meta-kaoline granules. Using the PMS, generated by pulp mill, as replacement for virgin fibers in production of fiber-cement sheets is already tested in pilot scale. The cost benefits analysis showed environmental and economic benefits for PMS generator as well as fiber-cement sheets producer (Modolo et al., 2011). Using the PMS for production of construction and insulation boards has been topic of The Waste & Resources Action Programme (Goroyias and Elias; 2004).

The use of the PMS as a paper and wood adhesive is an interesting concept under investigation (Geng et al., 2007). The use of PMS as cat litter is in production stage but without serious market demands. PMS can be dried in mixture with pesticides or fertilizers. Dried mixtures can be used as a pesticide or fertilizer carrier in agriculture which allows more controlled releasing active substances into the soil (Dongieux, 1999). Contamination associated with the sludge would be the ongoing threat in such a disposal route. Conversion of the lignocellulosic portion of PMS into methyltetrahydrofuran, which can be mixed with ethanol and natural gas into cleaner burning fuel, is already developed and has been found economically viable. Bio-conversion of PMS into ethanol is less viable due to toxic impurities in deinking sludges.

#### **5. Utilisation PMS for sorbent production**

Promising research has been conducted to use PMS as an oil sorbent material and use of the PMS as sorbent material is well documented, but currently the market was non receptive to such sorbent material due to cheap and efficient synthetic absorption material. The results of research studies have shown that PMS can be indirectly used as an active absorbent by converting it into activated carbon (Ben-Reuven, 2007). It can be used as binding material for the removal of heavy metals ions from water (Battaglia et al., 2003; Calace et al., 2003; Hea et al., 2010; Ahmaruzzaman, 2011), removal of phenols (Calace et al., 2002) and as an absorbent for hard surfaces cleaning (Lowe et al., 1988; Eifling and Ebbers, 2006). A variety of the processes and different absorbent products have been developed for commercial purposes. One of the processes which have been developed for the production of a floor absorbent, in the form of a granular product is known as the KAOFIN process and it is described in U.S.



Patent 4343751 (Naresh, 1980). In U.S. Patent 4374794 the sludge is evaporated, extruded into pellets and dried at temperatures ranging from 100 °C to 150 °C, in order to form an oil absorbent material (Kok, 1983). However, modern industry faces frequent and serious oil spills and subsequent sanitation demands high costs for sorbent materials. Offering a cheap and efficient natural material such PMS could become a welcome solution. The CAPS (Conversion of paper mill sludge into absorbent) is an eco-innovation solution in the 'market uptake' phase, therefore prior to expanding industrialization in Slovenia and Finland. The CAPS process uses the surpluses of the thermal energy which paper mills usually waste into the environment for sorbent production. In addition, CAPS uses paper mill waste as a secondary raw material and converts it into a high added value absorbent. The technology is relatively cheap, simple and easily replicable particularly in markets with a developed paper industry. It is based on drying of PMS to the point where it can be efficiently mechanically and/or chemically treated to release cellulosic fibers from its inorganic matrix. The humidity of the deinking and primary paper mill sludge lies between 50 to 70 %, whereas the content of cellulosic fibers is approx. 52 %, the remainder is inorganic. After drying between 70 - 80 % of the solid content, PMS proceeds through special mechanical treatment (unraveling). This stage is crucial for the entire process due to the fact that in this section cellulose fibers are released from the inorganic matrix, which in turn allows material to float on the fluid surface (Figure 5 and 6).

However, the mechanical treatment expanded the surface area, but the sorbency was not linear with regards to the surface area (Table 2).

	Avg. particle size mm	Active surface area m <sup>2</sup> /g	Sorbptivity g/g
Untreated PMS	10	4.8096	1.23
PMS Grinded	4.18	3.2048	2.07
PMS Unraveled	1.67	36.0526	4.4
PMS Fluffed	0.7	2.9626	7.12

Table 2. Particle size, active surface area and sorbency of used mineral oil versus mechanical treatment of PMS.



Fig. 5. Raw PMS (left), unraveled PMS (middle) and fluffed PMS (right).

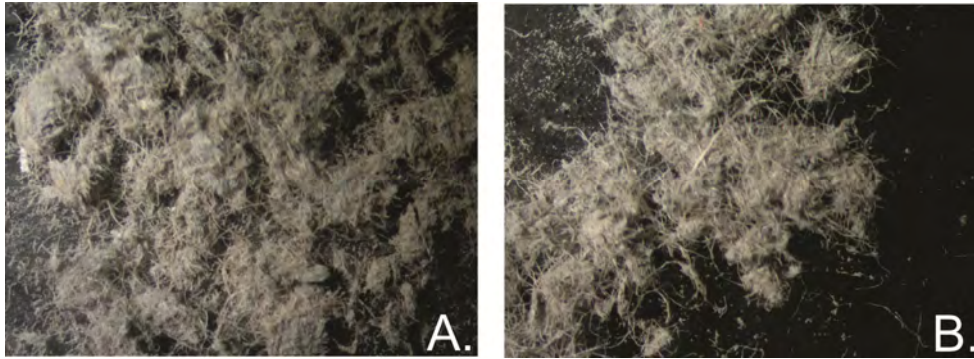


Fig. 6. Microscope pictures of raw PMS (A) and unraveled PMS (B) at 8 x magnification.

As can be seen from figure 7 the same trend was observed in sorbency of different substrates.

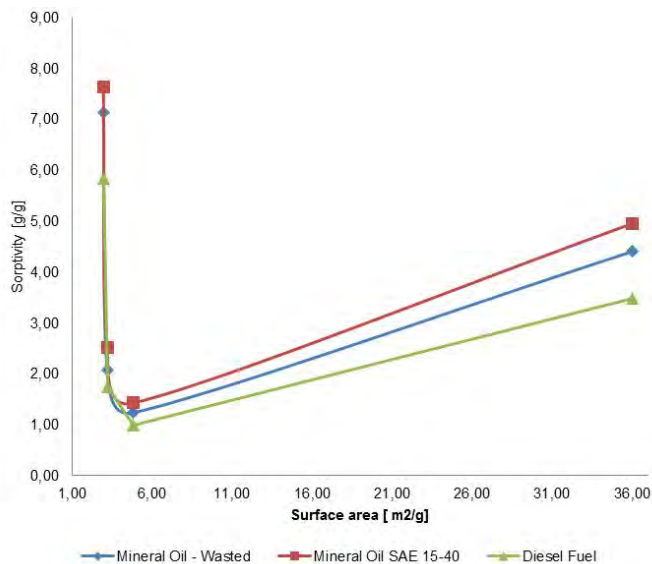


Fig. 7. Active surface area of PMS absorbent samples vs. mechanical treatment.

The mechanical treatment was clearly connected to the breaking of the inorganic matrix into dust. The dust dropped off from the absorbent, which led to the shrinking of the surface area but made the remainder of the surface more accessible to the substrate. More violent mechanical treatment during the production (fluffing) of the absorbent led the isothermal sorption of the absorbent to become similar to the isothermal sorption of the paper standard. The results showed that the appropriate treatment process (especially the mechanical treatment phase) was of the utmost importance for the conversion of the PMS into a sorbent. The possible explanation for this appears to be that the PMS consisted of a fragile net structure composed of cellulosic fibers and an inorganic core, which served as an anchor for linking the cellulosic fibers together. The inorganic core played an important role in the

sorption process because it served as absorption points for the hydrophobic substances, while the cellulosic fibers served as a floating skeleton. Chemical treatment (esterification, silanisation) is an option when higher absorption capacity and better buoyancy is required (Likon et al., 2011). Distributed (chemically prepared) paper mill sludge is dried at 130 °C to 150 °C until the humidity oscillates between 1% and 10% in order to obtain a final product with active surface area 36 m<sup>2</sup>/g, sorption capacity up to 8 g oil/g PMS and capacity to float on the water surface.

Produced sorbent has a calorific value around 3.8 MJ/kg and when it is completely soaked by oil it has a calorific value up to 33.5 MJ/kg and can be used as high quality fuel in the co-generation processes. Incineration under controlled conditions leads to the conversion of the kaolin portion of paper mill sludge into a meta-kaolin substance in the form of vitrified granules. These vitrified granules can be reused as an inert hydrophilic absorbent.

The adsorption kinetic shown (Figure 8), that the mechanism of PMS sorption follows complex combinations of interparticle diffusion at the first stage of the process followed by pseudo-second order adsorption of oil into pores of the inorganic part of PMS.

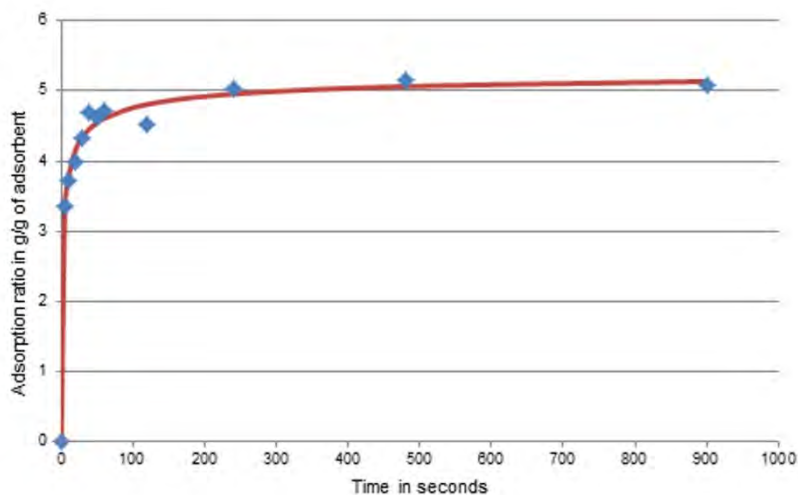


Fig. 8. Sorption kinetics curve for mineral motor oil (SAE 15-40W) sorbing on PMS at 20°C.

In the loose state of adsorbent the adsorption equilibrium is reached after 240 seconds at 20°C and as can we see from the figure 9 PMS absorbs more than 95 % of oil from a water surface within 70 seconds.

## 6. Environmental impact assessment of PMS sorbent versus expanded polypropylene sorbents

LCA model based on figure 10 showed that conversion of PMS from paper production can expand paper life cycle for additional step and helps closing the life cycle in paper production processes. A model presenting the life cycle assessments (LCA) of PMS sorbent



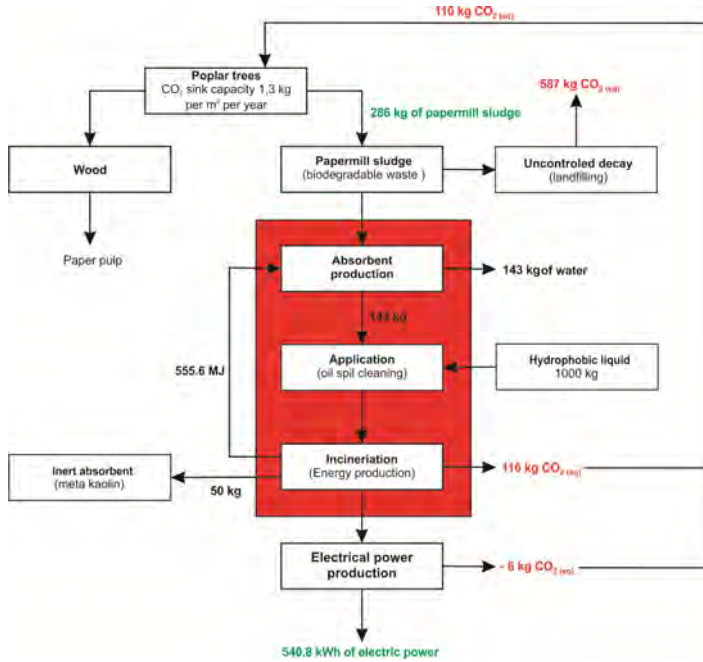


Fig. 11. Life cycle circle for conversion of PMS into absorbent material.

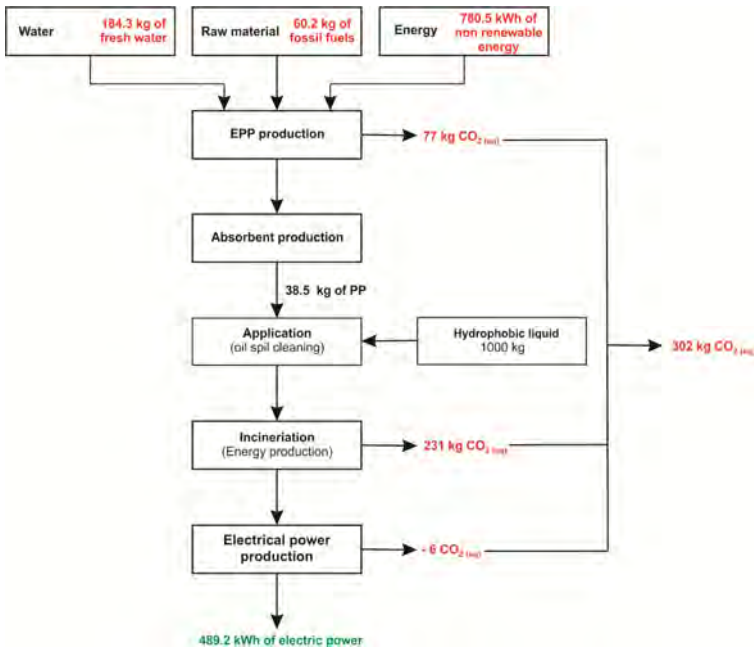


Fig. 12. Life cycle circle production and application of EPP absorbent.

The data for the uncontrolled anaerobic decay were calculated according to the equation for anaerobic decay (Buswell and Mueller, 1952) and the data for energy production were calculated according to Martin et al. (Martin et al., 2003). Paper mill sludge was considered as unwanted by-product in the paper and cardboard recycling processes that is without water and energy consumption and without GHG-s generation for its production. EPP was considered as absorbent produced from non-renewable raw materials (propene). The calculations were based on the quantity of absorbent required for the absorption of 1000 kg of oil with the following presumptions: the sorption capacity of absorbent produced from PMS and from EPP were 7 kg of oil/kg sorbent and 26 kg of oil/kg sorbent, respectively. It was considered that used sorbents were incinerated for energy recovery. Absorbed hydrophobic substances were not included in LCA calculations.

As can be seen from Figures 11 and 12, the production and application of PMS as a sorbent for oil spill sanitation reduced carbon footprint by 2.75 times when compared to the production and application of EPP. In addition, considering the production of absorbent instead of landfill disposal the PMS decreased carbon footprint by 5.25 times. Altogether, production of the sorbent material from PMS instead of landfill disposal it and replacing the synthetic EPP absorbent with PMS absorbent for oil spill sanitations reduced the carbon footprint by more than 14 times. The difference in the water balance was 372.3 kg calculated on the quantity of sorbent needed for sanitation of the 1000 kg of oil. The difference in the water balance was due to the consumption of the 184.3 kg of fresh water for EPP production and due to the production of 143 kg of clean technological water in the process of energy recuperation during PMS sorbent production. The energy balance showed the surplus of energy in the production of the PMS absorbent was due to the production of a combustible product from waste and negative energy balance in the case of the production and use of EPP absorbent. The later was due to heavy energy consumption during production of PP from fossil fuels. Additionally, the LCA analysis has shown that conversion of PMS into an absorbent prolongs life cycle of paper products for an additional two cycles and efficiently closes the life cycle circle of paper.

## 7. Conclusion

The modern sustainable management of production processes should be based on the industrial ecology approach, of which an essential element is the eco-symbiosis theory. Pulp and paper industry producing enormous quantities of solid waste what presents huge environmental burden. Appropriate managing with such a waste is most crucial task for modern pulp and paper industry. Many innovative approaches for conversion of the PMS into useful materials have been done in past two decades, but for many of them the markets demands have been too small for successful diverting of PMS from the landfill disposal. The CAPS process for the conversion of PMS into a sorbent for water surface cleaning fulfills all of above mentioned requirements. Moreover, it also includes the surpluses of thermal energy which paper mills usually transmit into the environment. The manufactured natural sorbent may be used by the oil, chemical, logistic and transport industries as well as public bodies such as fire brigades, civil protection and disaster relief institutions. These are institutions which all require an environmental friendly, efficient, cheap and at the same time sustainable product for cleaning of oil spills from water surfaces and/or for oil

separators maintenance and on the other hand the pulp and paper industry will expand their product portfolio by using waste as raw material. The main drawback of placing the PMS sorbent on the market is its slow degradation in water. That obstacle can be overcome by chemical treatment, proper application and by mixing PMS sorbent with floating materials but those processes raise the production costs and makes PMS absorbent unattractive to the market. Additional improvement must be made in the future to overcome water degradation of the PMS sorbent and with success the industry can obtain a sustainable, cheap sorbent for oil spill sanitation in unlimited quantities.

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