

# Review on Paper Waste Sludge and Feasibility of It and Its Components to Be Recycled in Polymer-Based Composites

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**Abstract**— Intervention into current building industry is the main strategy of European Commissions in order to achieve one of the targeted aims for sustainable development – to reduce the emission of greenhouse gases by 20% until 2020 and by 80% until 2050. Increasing requirements for building energy performance, which cover not only the improvement of thermal insulating properties of building materials but also the input of materials production and installation, force the sector of building materials to search for energetically efficient materials. The use waste is the most promising alternative for the production of building materials and their composites. This paper analysis one of the greatest waste – paper waste sludge, its physical and chemical composition, and feasibility to be recycled in building materials. The detailed review of paper waste sludge components – calcium carbonate, kaolinite and silica utilization for the production of polymer-based composites is presented.

**Index Terms**—Composites, Paper waste sludge, Recycling, Sustainable development.

## I. INTRODUCTION

On 25<sup>th</sup> of September, 2015 United Nations has drawn an agenda where 17 aims of sustainable development and 169 tasks for the implementation of the mentioned aims are foreseen. One of the aims intends to ensure sustainable models for consumption and production, i.e. to assure the management of harmful to the environment chemicals and waste, reduce their emission into air, water and soil until 2020 as well as the accumulation of waste by applying prevention, reduction, recycling and reuse [1].

Pollution prevention in paper production industry is the main worldwide aspiration [2]. One of the main aims of European Commissions and Directives – to transform European Union countries into “recycling community” [3–5] which is capable in reducing the accumulation of waste and using them in different processes [6].

Paper waste sludge (PWS) is the main and the greatest by-product in paper production (approx. 35% from the produced paper [7]), and its elimination as well as destruction is a primary environmental problem [8]. Nowadays, PWS is landfilled; however, increasing demands and consumption of humanity increases the production scale of paper as well as waste which intensively reduces the space in landfills. Concerning the legislations on the environmental pollution, increasing taxes and energy consumption arising from waste burning, landfills must be eliminated. Therefore, ways on how PWS could be used are extremely desirable.

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It is noticed that during well controlled burning process clayey materials existing in PWS, i.e. kaolinite may be transformed into metakaolin. Burnt PWS waste is characterised by high pozzolanic activity compared to industrial pozzolanic additives [9]; therefore, further feasibilities of PWS usage have been studied in the production of cement and ceramic products [10–11], structural [12–13], waterproof materials [14], lightweight structures [15–16] and absorbents [17] as well as agriculture [18–19] and synthesis of zeolites [20].

## II. LITERATURE REVIEW

### A. Paper Waste Sludge and Its Chemical Composition

Various solid waste and sludge form in different stages of pulp and paper production technological processes. The main source of it is waste water treatment. Solid waste and sludge arising from paper production may be classified into:

#### 1. Recycled paper waste.

1.1. Recycled paper waste consists of impurities, i.e. fibers, conglomerations, staples and metal pieces, sand, glass, plastic, scraps of paper fillers, materials for gluing and other chemicals. They are characterised by relatively low moisture content, are easily dried and in most cases burnt or landfilled.

1.2. Filter waste which form at the stage of pulp filtering when agglomerates of cellulose fibers which interfere production and worsen the quality of the final product are eliminated.

#### 2. Ink removal waste.

This waste consists of short fibers or particles, coating materials, fillers, ink particles (the source of heavy metals) and additives for the ink removal. For the separation of ink and fibers, floatation method is used where the resulting foams are gathered on the chamber surface of floatation equipment. Resulting ink removal sludge consists of ink, solid inorganic materials and short cellulose fibers.

#### 3. Primary sludge.

This sludge consists of fine particles and fillers which composition depends on recyclable paper. Primary sludge may be repeatedly used in the production of cardboard.

#### 4. Secondary sludge.

This sludge forms in waste water treatment equipment and may be recycled to cardboard products or pressed, dried and

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burnt or landfilled. The amount of secondary sludge is lower than primary sludge because most of it is removed from primary sludge as heavy, fibrous, inorganic and solid particles.

PWS is characterised by high moisture content (~50%), organic matter (wood, cellulose fibers which are too short for further recycling, lignin and in some cases organic binder (~25%)) and mineral charge comprising of calcium carbonate, kaolinite, talc and heavy metals (~25%); however, chemical composition and physical properties of PWS differ depending on the quality of paper, waste water treatment technology as well as specificity of fresh water usage [21]. Table 1 and Table 2 present the composition of PWS which has been determined by various authors. It can be observed that PWS consists of silica, aluminium, and calcium oxides.  $\text{SiO}_2$  or  $\text{Al}_2\text{O}_3$  are the main components of kaolinite and  $\text{CaO}$  – of calcium carbonate.

Author Bajpai 2015 has noticed that PWS particles are porous. Porosity (open or closed pores, the size and shape of pores) has a great influence on thermal insulating properties of building materials; therefore, the information on the use of PWS in thermal insulating materials or their composites is absent.

### B. Application of Paper Waste Sludge Components in Polymeric Systems

The application of such fillers as calcium carbonate ( $\text{CaCO}_3$ ) for the production of thermoplastics is well known practise in plastics industry when the price reduction of the final product is desirable. Properties of filled polymeric materials mostly depend on the shape, size and distribution of particles. There are five different shapes of particles – sphere, cube, flake, plate, fiber. The use of fiber-shaped particles increases tensile strength of composite and plate-shaped – rigidity which is mainly dependant on the aspect ratio of particles [36].

Scientists [37] have studied the impact of calcium carbonate particles on flexible polyurethane foam. The obtained results have shown that the addition of (100–250)  $\mu\text{m}$  sized  $\text{CaCO}_3$  particles when the amount varies from 1 wt.% to 30 wt.%, reduces even distribution due to overmuch varying particle sizes and increases the ability of particles to agglomerate due to uneven distribution of attraction forces between filler and polymer matrix. It is as well determined that a very high amount of calcium carbonate increases hysteresis and worsens the quality of the final product. For the production of flexible polyurethane foam, authors [38] have used fractioned particles of calcium carbonate (0.06  $\mu\text{m}$ , 0.5  $\mu\text{m}$ , 3.5  $\mu\text{m}$ , 10  $\mu\text{m}$ , 20  $\mu\text{m}$ , 841  $\mu\text{m}$ ) which amount has ranged from 5 wt.% to 40 wt.%. It is concluded that increasing amount of the filler by 75% reduces tensile strength independently of particles size. The use of too high amount of filler determines poor dispersion of particles and formation of agglomerates; wherefore, polymer matrix suffers from stress concentration. The similar reduction in tensile strength tendencies of flexible polyurethane foam modified with  $\text{CaCO}_3$  filler is observed by authors [39]. They have determined that optimal amount of calcium carbonate in flexible polyurethane foam is 20 wt.% based on the mechanical properties and production cost which is by 1.2 times lower than that of polyurethane foam without  $\text{CaCO}_3$

filler. Scientists [40] have noticed that flexible polyurethane foam without  $\text{CaCO}_3$  filler is distinguished oneself by structure consisting of uneven and large pores. Its formation is determined by homogeneous nucleation which requires higher activation energy. When filler is used, gases form at interface between filler and polymer matrix; they cause micro-cracks which lower activation energy necessary for the formation of polyurethane foam pores.

Authors [41] have observed that egg shells consist of high content of  $\text{CaCO}_3$  and decided to use this material as filler in polyurethane foam. The size of particles has ranged from 0.032 mm to 2 mm, and the amount – from 0 wt.% to 25 wt.% (step of 5 wt.%). The obtained products have had from 450 kPa to 430 kPa reducing compressive strength, from 1.2% to 1.5% increasing short-term water absorption as well as unaffected thermal (initial degradation temperature is 234°C, maximum temperature is 386°C) and dimensional stabilities.

The other component of PWS is kaolinite. Authors [42] have studied the impact of this and two other materials as fillers for polypropylene composites – talc (average size of particles is 6.3  $\mu\text{m}$ ) and calcium carbonate (average size of particles is 3.3  $\mu\text{m}$ ). They have determined that the addition calcium carbonate from 0 wt.% to 40 wt.% (step is 10 wt.%), has reduced tensile strength of composites from 18 MPa to 13 MPa, when the density is in the range from 850  $\text{kg/m}^3$  to 1140  $\text{kg/m}^3$ . It is assumed that it happens due to low interfacial adhesion between filler and polymer matrix which arises from insufficient wetting of filler particles. The addition of talc and kaolinite from 0 wt.% to 40 wt.% leads to insignificant increase in tensile strength, respectively, from 18 MPa to 24 MPa and from 18 MPa to 22 MPa when the density varies, respectively, from 850  $\text{kg/m}^3$  to 1200  $\text{kg/m}^3$  and from 960  $\text{kg/m}^3$  to 1190  $\text{kg/m}^3$ . Such increase in tensile strength appears because of strong interfacial adhesion which is simply obtained by using higher aspect ratio plate-shaped particles. Scientists [43] have studied the compressive strength of kaolinite modified composites and determined that the maximum value is obtained when 40 wt.% of kaolinite is added. Higher amount of this material determines the deterioration of mechanical properties due to reduced cross-link density arising from the greater distance between polymer chains and filler particles. Poly(methyl methacrylate) and polyurethane composites are characterised by similar compressive strength results obtained by [44] who has filled the products with 0.5  $\mu\text{m}$  sized kaolinite particles. He has determined that the increase in the amount from 0 wt.% to 5 wt.% increases compressive strength from 22.5 MPa to 36.0 MPa when the density is ranging, respectively, from 400  $\text{kg/m}^3$  to 1300  $\text{kg/m}^3$ , and the structure of composites distinguished oneself by smaller pores and higher pores density. Thermal stability results of such dense composites have shown that kaolinite acts as fire retardant and thermal stability increasing materials – initial decomposition temperature varies from 429°C to 504°C, and maximum temperature ranges from 444°C to 603°C, when the amount of kaolinite changes, respectively, from 0 wt.% to 5 wt.%. Basing on the density and strength results obtained by above mentioned authors, derived composite materials may be used for structural solutions.

Table 1: Chemical composition of PWS (a)

Chemical formula	Amount, wt.%						
	[22]	[23]	[24]	[25]	[26]	[27]	[28]
SiO <sub>2</sub>	32.6	44.0	30.2	25.1	21.1	21.6	16.7
Al <sub>2</sub> O <sub>3</sub>	27.3	29.2	18.0	18.5	13.6	14.4	16.5
CaO	27.1	4.2	31.4	33.6	37.8	36.5	22.5
MgO	7.1	7.8	2.7	1.2	0.5	2.4	-
Fe <sub>2</sub> O <sub>3</sub>	0.7	5.9	0.7	0.5	2.2	0.5	0.9
Na <sub>2</sub> O	0.1	0.8	0.2	0.2	-	0.1	0.3
K <sub>2</sub> O	0.2	0.3	0.3	0.3	-	0.4	-
SO <sub>3</sub>	-	-	0.2	-	-	0.3	-
TiO <sub>2</sub>	1.4	2.5	0.4	0.6	-	0.2	32.4
P <sub>2</sub> O <sub>5</sub>	0.9	0.3	-	0.2	-	-	0.8
LOI*	1.8	4.6	14.5	19.8	23.3	2.7	-

Table 2: Chemical composition of PWS (b)

Chemical formula	Amount, wt.%						
	[29]	[30]	[31]	[32]	[33]	[34]	[35]
SiO <sub>2</sub>	28.0	22.3	13.9	30.9	25.7	25.7	22.3
Al <sub>2</sub> O <sub>3</sub>	13.2	14.6	8.3	13.7	18.9	18.9	11.9
CaO	45.5	40.2	47.1	30.9	43.5	43.5	44.2
MgO	4.0	2.4	1.6	2.8	5.2	5.2	2.4
Fe <sub>2</sub> O <sub>3</sub>	1.3	0.6	0.5	2.5	0.9	0.9	0.6
Na <sub>2</sub> O	0.4	0.1	0.2	0.9	1.6	1.6	0.2
K <sub>2</sub> O	0.7	0.4	0.3	1.0	1.3	1.3	0.4
SO <sub>3</sub>	1.3	0.3	-	5.0	1.1	1.1	3.6
TiO <sub>2</sub>	0.7	0.3	0.3	-	0.7	0.7	0.4
P <sub>2</sub> O <sub>5</sub>	0.4	0.2	0.2	-	0.5	0.5	-
LOI*	5.7	18.5	26.7	8.3	-	1.2	3.3

\*) Loss on ignition.

Scientists [45] have analysed the possibility of silicate glass waste (particle size is in the range of (0.3–100) μm) to be used in building materials. Conducted X-ray analysis has shown that chemical composition of these waste is very similar to PWS, and thermal conductivity is 0.161 W/(m·K). The obtained results allow scientists drawing a conclusion that silicate glass waste may be used for the production of thermal insulating composite materials. Therefore, further research of authors [46] has been conducted on SiO<sub>2</sub> microparticles. They have determined that the added amount varying from 0 wt.% to 4 wt.% allows obtaining composites with compressive strength from 380 kPa to 430 kPa when the density ranges from 59.8 kg/m<sup>3</sup> to 72.5 kg/m<sup>3</sup> and reduces from 360 kPa to 270 kPa when the amount increases from 6 wt.% to 10 wt.%. Experimental results of water absorption have shown that SiO<sub>2</sub> microparticles determine the formation of closed cell structure; therefore, the addition from 0 wt.% to 10 wt.% into polymer system reduces water absorption from 1.3% to 0.60%.

### III. CONCLUSION

The conducted literature review has shown that increasing accumulation of waste requires additional measures. Therefore, landfilled paper production wastes should be additionally used for the production of structural or thermal insulating products. Chemical composition has shown that PWS consists of, mainly, calcium carbonate, kaolinite and silica which when used as fillers allow obtaining polymeric composite materials characterised by high mechanical

performance, sufficient thermal insulating properties and stability based on thermal degradation. The most impact on final product's properties having parameter is the shape of particles. Plate-shaped particles are the most suitable in order to obtain strong and rigid composites used for structural applications.

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## Review on Paper Waste Sludge and Feasibility of It and Its Components to Be Recycled in Polymer-Based Composites

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