The properties of wastepaper sludge ash and its generic applications

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Abstract. Wastepaper sludge ash (WSA) is generated during the recycling of paper, which is an industrial by-product can induce pollution to the environment. Due to its effect related to pollution, a generic review on its application is required. Despite many researchers conducted in the world to resolve the application of WSA as a sustainable material, the effects of WSA as a cementitious material in its application is still limited assess. Hence, this paper presents a generic review on the effect of WSA as a cementitious material. The generic review on WSA in term of its physical properties, chemical properties, reactive properties and application of WSA in the industry was carried out. From the review on WSA properties, it is found that the WSA has good potential as one of the important materials in the construction industry especially in the production of concrete, brick, mortar, soil stabilizing additive, rigid pavement and controlled low-strength materials (CLSM).

1. Introduction

Nowadays, there are many experiments conducted in order to replace the use of cement clinker with other materials such as waste ashes and industrial by-products. The waste ashes and industrial by-products are found contain pozzolanic properties and hydraulic activities similar to the properties of cement clinker which can be used as cementitious material (CM). Recently, several types of CM can be obtained in the industry such as fly ash, wastepaper sludge ash (WSA), ground granulated blast furnace slag and silica fume. Those materials are now widely used among CM. The materials have been tested and used for various applications. Advantageous of the use of these materials are beneficial for saving of natural resource, energy and reduction of CO2 emissions in the air which causes pollution to the environment.

Since there are many wastes have been produced and can be used as CM, in this paper, only WSA would be touch in-depth. WSA is one of the industrial by-products obtain from the paper mill and paper recycling industry. Normally, in the industry of papermaking, the waste paper has been burned and the CO2 has been emitted to the air. However, more than 70 % of WSA has been used in low-value application such as land dispersion. While the remaining 30 % of WSA has been thrown into dumpsites in the United Kingdom and annually produce approximately 125,000 tons of WSA [1]. The composition and properties of WSA vary according to the type of raw material whether it is WSA that is sprayed into the bed combustion unit or the combustion state. However, the WSA contains very high
alkaline with pH 12-13. It comprises primarily of calcium, silicon and aluminum oxide. Due to its compositions and properties, generic research on the use of WSA should be driven since it beneficial to be used as construction materials.

Numerous investigations on WSA have focused on hydraulic properties, pozzolanic reactivity and potential use as a cementitious material. It has been investigated by many researchers worldwide [2 – 18]. For instance, Chiang et al. [4], Weng et al. [10] and Liew et al. [11] have investigated the use of WSA as a lightweight building brick. The potential of reuse the wastewater sludge for innovative applications in construction aggregates has been investigated by Tay et al. [5] and Yagüe et al. [8]. Since the WSA can be used for various applications, its physical properties should be taken into account which it is able to give an input on the understanding on its generic applications.

2. Physical properties

As stated earlier, the WSA is produced by burning residual paper sludge from the papermaking process consisting of mineral fillers, small cellulose fibers, water, inorganic salts and organic compounds. Due to high air content of 40 % to 70 % in WSA, it would generate problems in term of handling, combustion and transmission. The residues must be firstly dried before being processed. Hence the physical properties of the WSA should be looked in depth. As stated by Trejo et al. [19], the amount and composition of the WSA vary depending on the resulting grade, the raw materials to be used, the processing techniques and the percentage of the WSA for its application. They add that it also depends on the characteristics of paper sludge. However, Mochizuki and Yoshino Etsuro Saito [20] added that the application of WSA on soil improvement is closely related to its composition and density of the particle. Where they found that the pH range of WSA extends from almost neutral to about 12, composition and density of the particles are about 2.2 – 2.9 g/cm³ and the maximum dry density is 0.65 – 0.95 g/cm³. According to Corinaldesi et al. [21], results from the experiment found that the water absorption capacity of about 25 % and specific gravity weight in the dry saturated state (SSD) was 1720 kg/m³. They also found that from 80 μm of paper sewage, the ash was 75 μm. From investigation performed by Monosi et al. [22] on the reuse of paper mill ash in plaster blends, found that the density of WSA is equal to 1200 kg/m³; lighter than ordinary sand, which has predicted density range from 2500 kg/m³ to 2600 kg/m³. They also found that the density of the binder is in the range of 2100 kg/m³ to 3100 kg/m³, which it is commonly used in mortar production. However, other researchers found that the average specific gravity of sludge ash was 2.81 [23] and 2.60 [24].

Investigation on sludge ash as fine aggregate in the concrete mix which conducted by Khanbilvardi et al. [25] found that the sludge ash is organic content which comprises of the water content of 28 % and a specific gravity of 1.83. Ishimoto et al. [26] noted that the paper sludge contains 60 % water and 40 % solids. And the solid contains 30 % ash, the other is losing during the flaming process. Liaw et al. [3] found that the average moisture content in the paper sludge was 75.40 % and ignition loss was 70.11 %. However, after the formation of the joint ash, the flame loss was found to be 19.63 %.

Mozaffari et al. [27] found that from the SEM spectrum, the WSA particles are porous and heterogeneous as shown in Figure 1. Due to its porosity, the particle of WSA seems to be agglomerated. It is due to the presence of irregular pores and fibrous nature in the WSA. The paper pulp holds the moisture in these pores and the fibrous envelopes providing an obstacle for moisture to move towards the surface. Balwaik and Raut [28] stated that the fibrous nature gives very high energy absorbing ability and hence produce high compressive strength. Segui et al. [29] added that due to the porosity of the WSA, it can create a problem in term of its workability. It is because the ash is a similar function like cement-based materials which is open porosity; hence it absorbs more water and leads to deteriorating its workability characteristics. Even though the physical properties of the WSA give information on its suitable application, the chemical properties of the WSA should also be touched in depth.
3. Chemical properties
The main components of WSA are calcium, alumina, silica, iron oxide, with varying carbon amounts, as measured by the loss in ignition (LOI). Summary of the chemical compositions of the WSA is presented in Table 1. Major constituents of the WSA are calcium (CaO), silica (SiO$_2$) and alumina (Al$_2$O$_3$). It also has a low percentage of magnesium oxide (MgO) and iron oxide (Fe$_2$O$_3$), which is higher than the minimum requirement set in OPC and ASTM C 618 (Class N). Sulphur trioxide, alkalis and LOI of WSA are lower than that of the upper limit set by ASTM. Therefore, the WSA is potentially used as an additive like Portland cement and also can be mixed with cement. Yan et al. [30] stated that sum of the oxides of Australian de-inking sludge from recycled waste paper is over 43% where it contains 23% of titanium oxide as the main component in the waste paper sludge. The waste paper sludge is also contained chloride ions of 600 ppm to 650 ppm [31 - 32]. It also contains metals such as copper, zinc, barium, lead and chromium (30 ppm to 300 ppm) [32 - 33]. Frias et al. [31 & 34] reclaimed that the chemical composition of the WSA from any given plant is highly consistent over time.

![Figure 1. SEM spectrum of WSA][28].

Table 1. Comparing the WSA’s with ASTM C618 [46] Class-N requirements and OPC.

| Chemical contain | OPC [35] | [36] | [37] | [38] | [39] [27, 40] | [41] | [42] | [43] | [34] | [44] | [30] | [45] | ASTM C618 [46] |
|------------------|---------|-------|-------|-------|--------------|------|------|------|------|------|------|------|--------------|
| SiO$_2$          | 18.7    | 44    | 32.6  | 32.55 | 25.7         | 25.7 | 28   | 21.9 | 18.05 | 10.79 | 10.69 | 6.97 | 6.42 |
| Al$_2$O$_3$      | 2.2-6.3 | 29.2  | 27.3  | 14.13 | 18.9         | 18.9 | 13.2 | 11.2 | 10.14 | 6.82  | 6.74  | 6.9  | 4.14 |
| Fe$_2$O$_3$      | 0.2-6.1 | 5.9   | 0.7   | 1.76  | 0.9          | 0.9  | 1.3  | 0.8  | 0.55  | 0.46  | 0.41  | 0.38 | 0.28 |
| $^{4}$SAF        | 79.1    | 60.6  | 48.44 | 45.5  | 45.5         | 45.5 | 35.9 | 28.74| 18.07 | 17.84 | 14.25 | 10.8 | 4   |
| SAF + Al$_2$O$_3$+Fe$_2$O$_3$ | SAF$^a$ | N=70  | min  |       |              |      |      |      |      |      |      |      | 32  |
| CaO              | 60.2-68.7 | 4.2  | 27.1  | 37.85 | 43.5         | 43.5 | 45.5 | 14.3 | 19.82 | 25.43 | 24.15 | 9.37 | -   |
| MgO              | 0.3-4.8 | 7.8   | 7.1   | 5.47  | 5.2          | 5.2  | 4    | 4.1  | 2.58  | 0.86  | 0.26  | 0.11 | 1.54 |
| SO$_3$           | 1.7-4.6 | -     | -     | 1.1   | 1.3          | 1.3  | -    | -    | 0.33  | 0.33  | 0.3  | -   | 10 max |
| LOI              | -0.3    | 4.6   | 1.8   | -     | 1.2          | 0.5  | 5.7  | 46   | 47.62 | 54.34 | 55.71 | 58.8 | 53.8 |

Moreover, the WSA also contains a small number of minerals such as kaolinite and calcium carbonate, which normally used as coating agents to ensure smooth paper surfaces. Typically, the coating agent of 5 g/m$^2$ to 20 g/m$^2$ is depending on its quantity and paper type. The minerals that generally in the WSA are gehlenite (Ca$_2$Al$_2$SiO$_7$), lime (CaO), calcite (CaCO$_3$), quartz (SiO$_2$), merwinite (Ca$_3$Mg(SiO$_4$)$_2$) and $\alpha'$-Ca$_2$SiO$_4$. Bai et al. [39] found that Gehlenite is a major mineral and gives a strong fission pattern when combined with lime. An assumption has been made that the presence of bredigit (Ca$_{14}$Mg$_{6}$(-SiO$_{38}$)) is indistinguishable from the presence of $\alpha'$-Ca$_2$SiO$_4$ due to both minerals have parallel XRD patterns. Studies carried out by Villa et al. [47], Frías et al. [48] and...
Garcia et al. [49] found that the XRD analysis results are very important for WSA generators as a hydraulic binder since the lime (CaO), mayenite (Ca\textsubscript{12}Al\textsubscript{14}O\textsubscript{33}) and \(\alpha'\)-Ca\textsubscript{2}SiO\textsubscript{4} are reactive minerals in the WSA.

The metal aluminum (Al) element is also found in the WSA. It is normally contained in about 0.2 % to 0.3 % as it is often detected in waste from municipal solid waste combustion. The metallic aluminum causes swelling if the WSA is introduced into cement-based material. This is due to the reaction between Al and water with the presence of alkali in cement. The reaction releases hydrogen, which causes expansion through the determination of cement-based material containing WSA, is indicated in Equation 1 [41].

\[
2\text{Al} + 2\text{OH}^- + 2\text{H}_2\text{O} \rightarrow 2\text{AlO}_2^- + 3\text{H}_2
\] (1)

From the review on chemical properties found that the WSA is containing a lot of compositions which affect to its application. Other important properties that play important roles on the determination of its application are reactive properties.

4. Reactive properties

The WSA is also contained the reactive properties and many investigations have been performed by many researchers [42, 47, 50 - 51]. The temperature effect is also been investigated. Many researchers [42, 47, 50 - 51] found that 700 °C to 750 °C induces the formation of reactive pozzolanic material in WSA. The presence of CaO in the WSA as a hydraulic binder has negative and positive concerns for its extensive use [39]. This is because, enlargement of CaO hydration to Ca(OH)\textsubscript{2}, which results in uncertainty after setting. Boni et al. [52] found that WSA does not symbolize the major hazards of the environment in terms of the release of heavy metals.

The similarity between properties of WSA and cement can be achieved if the WSA reacts with water, sets and hardens. However, the WSA is very high-water demand due to its high porosity. At the same time, the existence of Al in the WSA is also reported to cause swelling and expansion under alkali conditions. Cracking caused by shrinkage is also another potential problem. Nonetheless, the stirring of WSA with broken sand degradable clay increases the development of strength and reduces some of the adverse effects [27 & 39]. However, it is found that small surges in compressive strength can be achieved in mortar specimen when 5 % of cement weight is replaced with WSA [53]. Hence, the reactive properties in WSA are found useful information in order to identify its suitable application. The WSA can be used as a promising material to be used in civil engineering construction applications since some investigation found that the compressive strength of the specimen can be achieved with a certain level of WSA.

5. WSA in civil engineering construction applications

There are many benefits can be obtained by utilizing the WSA in civil engineering construction application. The advantageous are can reduce the cost of disposal and less reserved area for disposal (thereby enabling other land uses and release emissions that permit requirements). Moreover, from the sale of by-product or at least offset processing and disposal costs, it is considering as cost-effective. Summary of its application based on the percentage of replacement, adopted material application and outcome is presented in Table 2. From Table 2 found that the WSA can be mixed in the application of concrete, mortar, brick, soil stabilizing additive, rigid pavement and controlled low-strength materials (CLSM). Those applications are several parts of material production that generally used in civil engineering construction.

6. Conclusion

The physical properties, chemical properties and reactive properties of the WSA have been reported. In sum, it is found that the WSA has good potential as one of the important materials in the civil engineering construction industry especially in the production of concrete, brick, mortar, soil stabilizing additive, rigid pavement and CLSM.
Table 2. Key applications identified to sustain existing growth in the utilization.

| Author(s) | % of replacement | Adopted application | Outcomes |
|-----------|------------------|---------------------|----------|
| [54–64]  | 0–40%            | Concrete & Foamed Concrete | • The replacement of WSA shows a positive effect with 5–10% ↑ the mechanical performance (compressive strength and splitting tensile strength and flexural tensile strength) by weight and with 30% ↑ its start ↓ in strength.  
  • ↑ fineness and consequently ↑ water absorption, it required a ↑ amount of water.  
  • The use of WSA should not be ↑ than 10% by weight of the cement replaced unless mortar mixtures are judiciously proportioned.  
  • The replacement does not exceed 10% WSA yield ↓ values of mechanical strength resistance, incompatible with market requirements for brick production at a firing temperature of 900 °C.  
  • The ↓ heat of hydration of the waste material which generates ↓ reactions.  
  • The optimum firing temperature was 950 °C. The thermal conductivity ↓ with ↑ in percentage of deinking paper mill sludge.  
  • 15% deinking paper mill sludge gives the optimum strength at a firing temperature of 950°C.
| [65–67]  | 0–30%            | Mortars Brick | • The optimum concentration of WSA to stabilize the peat soil is about 7–20% of the maximum compressive strength from 0 days to 28 days of curing periods.  
  • The use of WSA demonstrates the advantages of technology, economics and environmental advantages in the stabilization of sulphate soils and other clay, as an alternative to the traditional stabilizers of chalk and or Portland Cement.  
  • With the addition of 10% WSA, the California Bearing Ratio (CBR) values have been fixed for stable soil for soaked and non-invasive conditions compared to control (unstable clay).  
  • ↓ reactions.  
  • The optimum firing temperature was 950 °C. The thermal conductivity ↓ with ↑ in percentage of deinking paper mill sludge.  
  • 15% deinking paper mill sludge gives the optimum strength at a firing temperature of 950°C.
| [68–72]  | 2–30%            | Soil stabilizing additive | • The optimum concentration of WSA to stabilize the peat soil is about 7–20% of the maximum compressive strength from 0 days to 28 days of curing periods.  
  • The use of WSA demonstrates the advantages of technology, economics and environmental advantages in the stabilization of sulphate soils and other clay, as an alternative to the traditional stabilizers of chalk and or Portland Cement.  
  • With the addition of 10% WSA, the California Bearing Ratio (CBR) values have been fixed for stable soil for soaked and non-invasive conditions compared to control (unstable clay).  
  • ↓ reactions.  
  • The optimum firing temperature was 950 °C. The thermal conductivity ↓ with ↑ in percentage of deinking paper mill sludge.  
  • 15% deinking paper mill sludge gives the optimum strength at a firing temperature of 950°C.
| [73–74]  | 0–40%            | Rigid Pavement | • 2% CBR value and wheel load (P) of 30kN; Rigid pavement costs ↓ by 73.83% (relative cost).  
  • 30% of WSA content expressed as a large percentage of recycled concrete aggregate (RCA) is to produce a uniform mix with continuous ↑ strength.
| [75–76]  | 5–30%            | CLSM | • 2% CBR value and wheel load (P) of 30kN; Rigid pavement costs ↓ by 73.83% (relative cost).  
  • 30% of WSA content expressed as a large percentage of recycled concrete aggregate (RCA) is to produce a uniform mix with continuous ↑ strength.

↑ (Increased/High); ↓ (Decreased/Low)

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