Progress in manufacture and properties of construction materials incorporating water treatment sludge: a review

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Abstract

Water treatment sludge (WTS) management is a growing global problem for water treatment plants (WTPs) and governments. Considering the scarcity of raw materials in many parts of the planet and unique properties of WTS, extensive research has been conducted on the application of WTS in the production of construction materials such as roof tiles, bricks, lightweight aggregates, cement, concrete and geopolymers. This paper critically reviews the progress in the application of WTS in construction materials, by synthesizing results from recent studies. Research findings have revealed that incorporation of $\leq$ 10% alum-based sludge in ceramic bricks is satisfactory with a small reduction of mechanical performance. Using the iron-based sludge, the bricks presented better mechanical strength than the reference clay-bricks. Concerning WTS application in concrete, 5% replacement of cement or sand by WTS was considered as the ideal value for the application in a variety of structural and non-structural concrete without adverse effect on concrete mechanical performance. Furthermore, this paper discusses sludge-amended concrete in terms of durability, potential leaching of toxic elements and cost, and suggests topics for future research on the sustainable management of WTS.

Keywords: Water treatment sludge; Construction materials; Mechanical properties; Durability; Solid waste recycling

1. Introduction

To protect public health, raw water is treated to a high standard at water treatment plants (WTPs) before drinking water distribution, through the process of coagulation, flocculation, sedimentation, filtration and disinfection (Ahmad et al., 2016; 2017). Large quantities of water treatment sludge (WTS) are produced daily by WTPs, and its management is becoming a global problem of major concern (Dassanayake et al., 2015; Ahmad et al., 2016; Hidalgo et al., 2017; Lee et al., 2018). A recent survey by Maiden et al. (2015) showed that the amount of WTS generated by Australian WTP could reach up to 43500 tons per year. In 2011, Japan generated over 290000 dry tons of sludge across the country (Fujiwara, 2011), while the UK produced around 131000 tons in 2014 (Finlay, 2015). **Figure 1** shows an estimated amount of sludge per capita produced in selected countries. It is estimated that an ordinary WTP generates over 10000 tons/day and 100000 tons/year of WTS (Babatunde and Zhao, 2007; Ahmad et al., 2016).

A major environmental challenge for water treatment is the disposal of excessive sludge produced in the process, which are either discharged into waterways or disposed to landfills (Ooi et al., 2018). The raw water quality and the treatment process are the key factors determining the quantity and quality of the sludge produced. Thus, any change in the quality of natural water, seasonal change, as well as the dosage of the coagulants used in the treatment system, will alter the quantity and quality of the sludge produced (Ahmad et al., 2016).

Curiously, in the scientific literature, there are limited statistical data on the production and costs associated with WTS on a global or national scale (Babatunde and Zhao, 2007; Keeley et al., 2014; Ahmad et al., 2016). Furthermore, other problems such as the risk of water contamination with heavy metals, high costs of sludge treatment and disposal are challenges faced by water companies and governments around the world. In Australia, the cost associated with the sludge disposal without transportation is $130-200/ton through landfill or/and sewer disposal, with an approximate cost of over $6.2 million per annum just in the state of Victoria (Maiden et al., 2015). The costs associated with sludge disposal in the Netherlands are on average between £30 and £40 million (Evuti and Lawal, 2011), while in the UK the annual disposal costs £5.5 million (Keeley et al., 2014).

WTS has been used in various industrial and commercial manufacturing processes. Particularly, some studies have been conducted by using WTS for contaminant removal in wastewater treatment, coagulant recovery, in ceramic products, and land application (Li et al., 2013; Ahmad et al., 2016; Cremades et al., 2018; Hagemann et al., 2019). As clay and WTS have a similar mineralogical composition (essentially hydroxides and oxides of silica, aluminum and ferric), the use of WTS has been highly encouraged to partially replace the clay used for fabrication of cement and others sintered ceramic materials (Cremades et al., 2018; Hagemann et al., 2019). Furthermore, because of clay scarcity in many parts of the world and the high environmental impact generated by the clay harvesting, many studies are focused on designing and development of new sustainable materials using less conventional raw materials, with available local sources and in accordance with international standards requirements. In order to protect the natural environment and the limited clay resources, countries such as China have begun to limit the use of clay-bricks and publicly recommend the incorporation of waste materials into the ceramic products (Chen et al., 2011).

There are a few review papers on WTS application and reuse (Babatunde and Zhao, 2007; Raut et al., 2011; Ahmad et al., 2016), most of which have focused on general aspects of brick or cement production from the last decade. Furthermore, the majority of the studies do not differentiate between the different types of sludge such as iron-based, aluminum-based, or lime (calcium carbonate, $CaCO_3$) which have been applied in the building materials, yet they have different properties and behaviour. This review therefore evaluates the different types of WTS and their latest applications in building materials such as ceramic bricks, paving tiles, lightweight aggregates, cement, concrete and composite materials. In addition, it discusses important aspects such as durability, environmental risks and cost-benefit analysis from the application of different types of WTS. Recommendations are provided for future studies in this emerging research topic.

2. Characteristics of water treatment sludge

2.1. Chemical properties

In most water treatment systems around the world with elevated suspended particles, inorganic coagulants are more commonly applied (Kashyap and Datta, 2017). They are composed mainly of iron and aluminum salts such as aluminum sulfate [$Al_2(SO_4)_3$], ferric sulfate [$Fe_2(SO_4)_3$] and ferric chloride ($FeCl_3$). Whereas in the softening process with high levels of hardness, the main coagulant used is lime (Sales et al., 2011). These coagulants are effective in removing a wide variety of impurities from the water, including colloidal particles and dissolved organic substances. The final process of water filtration results in a range of by-products known as WTS, which consists of a mixture of microorganisms, organic suspended compounds, coagulant and chemical oxides (Babatunde and Zhao, 2007; Ahmad et al., 2016). The final composition of WTS is the result of the coagulant type and other chemical treatment used for the purification of water. The typical sludge composition is shown in **Table A1**.

Particularly, aluminum sulfate has been used worldwide for more than 100 years in different WTPs, due to its low cost, easy manipulation and storage (Renault et al., 2009). Most WTPs in New Zealand and Australia use aluminum-based coagulants (Ministry of Health, 2017). However, in the most populated NSW State in Australia iron-based coagulants are used in the majority of its WTPs due to severe manganese limits in the finished water. The United Kingdom uses an average of 107000 tons of aluminum-based coagulant and 165000 tons of iron-based coagulants per year at WTPs across the country (Henderson et al., 2009). In 1994, in the United States and Canada, 72% of all water produced came from coagulation with aluminum salts and 23% with iron salts. There are several factors that lead to the use of one or another coagulant in water treatment, including the search for better quality of treated water, local availability, lower cost, smaller volume of sludge and best dehydration conditions (Pizzi, 2010).

2.2. Geotechnical properties

The knowledge of WTS geotechnical properties is essential to facilitate its application (Keeley et al., 2014). The properties such as mass loss on ignition (LOI), liquid limit (LL), plastic limit (PL), plasticity index (PI) and specific gravity are crucial not only for WTS identification or classification as a soil material, but also to predict future behavior of the application of this material (Komlos et al., 2013; O'Kelly, 2008). **Table 1** shows that the majority of geotechnical characterization studies with WTS have demonstrated a substantial similarity with clayey soils, especially WTS from aluminum and iron salts. However, WTS is different to clays due to the presence of organic matter and high concentrations of chemicals (Lee et al., 2018). Such organic matter in the soil usually results in high plasticity and low permeability, and decreases the specific gravity (Mitchell and Soga, 2005).

O'Kelly (2008) analyzed the geotechnical properties of WTP residuals that had aluminum as a coagulant. The study characterized the sludge as a clay with high plasticity, high compressibility, and very low permeability, which were attributed to the high affinity of the coagulant metal by water and the high content of organic particles. These characteristics of natural and untreated sludge make it sometimes inappropriate for applications in building materials such as aggregates and structural elements, and help to explain the difficulty of handling and transporting such material. Thus, by proper treatment to the sludge and its incorporation into the most suitable material, the negative aspects of these characteristics can be mitigated (Keeley et al., 2014).

3. Application of water treatment sludge in construction materials

3.1. Bricks and ceramic products

Considering the vast potential of solid waste from various industries as clay replacement for the production of ceramic artefacts (roof tiles and bricks), the latest research has focused on offering alternatives and environmentally friendly destination for these by-products. In addition, materials resulting from the sintered ceramics are tolerant of additions of other raw materials even in high quantities, promoting the solidification and immobilization of the elements with toxic potential (Dondi et al., 2016; Jonker and Potgieter, 2005) and high durability (Toya et al., 2007). Therefore, due to the fact that clay and WTS have a similar chemical composition (essentially hydroxides and oxides of silica, aluminum and ferric), the use of WTS has been highly encouraged to partially substitute the clay used for fabrication of bricks and ceramic materials (Toya et al., 2007; Cremades et al., 2018). This is especially the case in countries such as Brazil, where the production of sintered construction materials (bricks, tiles, blocks) is still more advantageous than cement and geopolymeric materials which will remain so for the next two decades (Monteiro and Vieira, 2014).

The amount of sludge added in the ceramic materials depends partly on its property (particle size, chemical and mineral composition), which will directly influence the quality of the bricks produced (Teixeira et al., 2011). In most studies, the increase in the addition of sludge in the brick mix resulted in a decrease in compressive strength and higher water absorption. The high values of loss on ignition are related to the high concentration of organic matter contained in the sludge, which was burned during the sintering process, combined with the presence of inorganic components found in WTS and clay (Wolff et al., 2015).

Teixeira et al. (2011) showed that aluminium-based sludge was more deleterious than iron-based coagulants to ceramic materials. In general, the iron-based sludge applied in ceramic products obtained better results concerning the mechanical properties and also a reduction in the firing temperature of bricks (Anderson et al., 2003; Teixeira et al., 2011; Kizinievič et al., 2013). Furthermore, Kizinievič et al. (2013) concluded that usually the iron-sludge conferred a more reddish colour to the bricks, acting as a natural pigment. However, the results with the addition of high proportions of aluminium-based sludge show a considerable reduction in the mechanical performance of the ceramic bodies with increasing sludge addition (Huang et al., 2001; Teixeira et al., 2011). In contrast, Benlalla et al. (2015) obtained bricks with mechanical properties higher than reference clay bricks, even with the substitution of clay by around 30% WTS at a firing temperature of 1000 °C.

A significant amount of studies discussing the use of WTS in combination with other alternative materials have been reported in **Table A2**. In a UK study, Anderson et al. (2003) investigated the incorporation of two by-products in the manufacture of bricks. They used sludge generated during water and sewage treatment, which was incinerated and added as partial substitutes for the traditional raw materials of the brick, in proportions of 5% (Dry weight), meeting local standards and parameters for ceramic blocks. In a similar study in Taiwan, Huang et al. (2001) mixed the sludge from the water treatment with dam sediment for the production of ceramic samples and showed that WTS (< 20%) combined with dam sediment can generate quality bricks under Chinese National Standard. In Taiwan, Chiang et al. (2009; 2010) produced lightweight bricks by mixing dry WTS and calcined rice husk, which showed improvements in mechanical properties. In addition, several other studies have demonstrated successful application of waste materials including WTS with a high concentration of silica in making ceramics and bricks with good mechanical performance (Hegazy et al., 2012; Wolff et al., 2015; Ewais et al., 2017).

In general, the addition of aluminum-based sludge decreases compressive/flexural strength and increases the water absorption of the ceramic bodies. According to the majority of the studies summarized in **Table A2** (Huang et al., 2001; Teixeira et al., 2011), the replacement of clay by aluminum-based sludge below 10% did not significantly reduce the mechanical properties of the ceramic bodies, although above 10% a reduction of 24.6-45.45% in flexural strength was reported. On the other hand, iron-based sludge increased the compressive/flexural strength of the ceramic samples by 7-97% compared to the reference values, for sludge addition below 10% (Kizinievič et al., 2013; Hassan et al., 2014). Furthermore, studies have shown that it is possible to produce ceramic bricks with mechanical properties similar to those of clay, with more considerable additions (> 50%) of sludge, combined with material with high silica concentration (Lin and Lin, 2005; Chiang et al., 2009; Hegazy et al., 2012; Wolff et al., 2015; Ewais et al., 2017).

3.2. Lightweight aggregates

Lightweight aggregates (LWA) are highly porous and spherical ceramic products with low density (0.8-2.0 g/cm^3) and commonly used in the manufacture of various building products (Soltan et al., 2016). **Figure A1** shows the manufacturing process of traditional LWA from clay. The use of residual waste materials in the production of LWA has been extensively exploited in recent years, mainly because LWA has wide applications in construction materials such as lightweight concrete/mortar (structural and non-structural), water treatment process and gardening (Dondi et al., 2016). A significant number of studies with sludge and industrial waste have used the thermal stabilization process to produce composites LWA, immobilizing the contaminants e.g. heavy metals through the sintering process in the matrix of the new materials, and thus reducing the migration of contaminants into the environment (Huang et al., 2007; Wang et al., 2008; Corrochano et al., 2011; González-Corrochano et al., 2017).

3.2.1. Lightweight coarse aggregate

Huang et al. (2005) used the aluminum-based sludge from a Taiwanese WTP to produce lightweight concrete coarse aggregates. Spherical samples of approximately 2 cm in diameter were produced, which were oven dried at 105 °C for 24 h and sintered at 1000, 1050 and 1100 °C. The WTS-derived LWA had water absorption and specific gravity of 14.47-37% and 1.12-1.78 g/cm^3, respectively. After composite production, the LWA was applied to a concrete mix with proportions of 2:1:1 (Natural sand: WTS-LWA: cement). The results, when compared to the Taiwanese construction standard, showed that only the WTS-aggregates sintered at 1100

°C met the requirements of specific gravity (1.4-2.0 g/cm^3) and compressive strength (175-420 kgf/cm^2) of the concretes made with natural LWA.

Huang and Wang (2013) investigated the alum-based sludge from 10 WTPs in Taiwan to produce lightweight coarse aggregates (**Figure A2**). By using air-dry sludge mixed with an adequate amount of water which was then extruded to produce cylindrical pellets of 8-12 mm in diameter, they obtained samples which were sintered at 500 °C for 5-15min and then 1150-1275 °C for 7.5-15 min. They reported satisfactory results with LWA concrete made with WTS, five were classified as viable for structural and non-structural LWA production, the other five only for structural, all with similar properties of high-quality LWA, with densities of 0.65-2.05 g/cm^3 and water absorption of 0.5-15%. Moreover, the researchers produced LWA in large scale using a rotary kiln with sludge from one of the WTPs. The results demonstrated that it was possible to produce structural and non-structural LWA with average particle density of 1.35 g/cm^3 and bulk density of 726 kg/m^3 meeting the requirements of the American Society for Testing and Materials (ASTM, 2017b).

In a different approach, Sales et al. (2010; 2011) produced a composite lightweight coarse aggregate for concrete with alum-based WTS and sawdust. For the lightweight composite production, dry and milled WTS was homogenized with water and sawdust, in a ratio of 6: 4.5: 1, to generate rounded samples with a diameter of 14±2 mm. After molding, the samples were dried at 105 °C and then immersed in boiled linseed oil for 1 min. Then the samples were dried at room temperature and mixed in different concretes batches. Based on the ACI (2014) classification, the concrete produced with the lightweight composite was in the non-structural lightweight concrete category, showing an axial compression strength of 11.1 MPa and an apparent density of 1847 kg/m^3.

3.2.2. Ceramsite

Ceramsite is a type of fine LWA that has been used as construction materials in a variety of applications, such as concrete composites, bricks, wetlands layers and filters for wastewater treatment process (Xu et al., 2008b; Jia et al., 2014). Xu et al. (2008a) tested the feasibility of ceramsite production with alum-based WTS and wastewater treatment sludge (WWTS). For the manufacture of ceramsite, different proportions of WTS, WWTS and water glass (sodium silicate) were used to obtain pelletized samples (5-8 mm) which were maintained at about 20 °C for 3 days and then pre-heated to 24 h at 110 °C. The heating ramp started at 20 °C and continued at a rate of 8 °C/min in a muffle until reaching 1000 °C for 35 min. The results showed that the optimal values for the manufacture of ceramsite were: WTS/WWTS = 45/55, sodium silicate/(WTS+WWTS) = 20%, SiO_2 = 14-26%, and Al_2O_3 = 22.5-45%, at a sintering temperature of 1000 °C. Under these conditions, the compressive strength of ceramsite ranged from 13.63 MPa to 16:25 MPa, above the minimum (7.50 MPa) required by the National Standards in China. In a similar research, Zou et al. (2009) investigated the effect of different proportions of Fe_2O_3, CaO and MgO oxides on the production of ceramsite with WTS and WWTS. The methodology used was the same as used by Xu et al. (2008a), however varying and adjusting the amount of iron, calcium, and magnesium oxide. The variation of Fe_2O_3 content directly influenced the mechanical strengths of ceramsite mixtures. Fe content of 5-8% was considered optimal for lower water absorption and higher compressive strength which ranged from 14.97 MPa to 15.67 MPa. The mechanical strength of ceramsite was not influenced directly by MgO, but an increase in the bulk density was noticed as MgO was increased, resulting in an ideal rate of 1.6-4.0%. The increase in CaO amount decreased the compressive strength of ceramsites, and an optimal value of 2.75-7% was proposed.

3.3. Cement production

Global Portland cement and concrete production (**Figure A3**) is a technological activity of intense demand for natural resources. The raw materials commonly used in the manufacture of cement are calcium carbonate (75-80 wt.%) and clay (20-25 wt.%) (Bignozzi, 2011). Due to the mineral and chemical similarity of Portland cement and WTS, alternatives are being sought to incorporate WTS into cement production process or as a supplementary cementitious material.

Tay and Show (1991) used an iron-based WTS mixed with different proportions of lime powder ($CaCO_3$) and sintered at a temperature of 1000 °C for 4 h in three different blends (1:3, 1:1 and 3:1 by weight). The results indicate that cement made from Fe-sludge, with a sludge-lime mixing ratio of 1:1, sintered at 1000 °C for 4 h, can be used for general masonry work according to ASTM. On an industrial scale, Baker et al. (2005) replaced 15% of limestone for lime sludge (Ca-sludge) in a cement plant and produced around 80 tons of composite cement. The properties of the new cement were satisfactory in comparison to the ordinary Portland cement. Similarly, Pan et al. (2004) evaluated the possibility of replacing clay used in Portland cement clinker production by using alum-sludge. The cement containing WTS as a substitute of clay showed adequate results regarding the setting time of cement paste. The compressive strength of concrete was increased as the sludge was incorporated, meeting the Chinese national standard requirements for first grade Portland cement.

Lin and Lin (2005) used WTS ash combined with two more industrial by-products for replacement of Portland cement raw materials. The results regarding compressive strength and microstructure of cement paste showed that the substitution up to 20% of these materials was feasible for the manufacture of clinker cement. Moreover, all hydrated compounds commonly found in ordinary Portland cement paste [$Ca(OH)_2$ and C-S-H] were detected after the addition of these residues. Chen et al. (2010) studied the feasibility of replacing the siliceous raw material (Shale) by using alum-sludge from 4% to 10% for cement production. All specimens with WTS had higher 3-day and 7-day strength than the control specimens, and also superior percentage of tricalcium silicate (C_3S). However, after 28-day only samples with sludge additions below 7% had higher strengths than the control. In more recent studies, Dahhou et al. (2016; 2018) produced several clinker compositions with partial limestone replacements by alum-sludge and sintering temperature at 1300-1500 °C. Regarding the flexural and compressive strength of the cement pastes burned from 1450 and 1500 °C, all clinker compositions showed superior performance and also identical mineralogical values compared to ordinary Portland clinker, with a significant presence of alite (C_3S) and belite (C_2S).

The production of cement with the partial addition of WTS has been shown to be potentially feasible. Most of the studies focused on cement paste parameters such as mechanical strength, setting time, chemical and mineralogical characterization and some leaching tests. However, due to the lack of microstructure and long-term durability testings on concrete and the presence of some substances in sludge such as chloride and sulfate ions which are potentially deleterious to concrete, it is unclear how the use of WTS will affect the microstructure and long-term performance of concrete. For example, in this case whether the expansive products (such as ettringite) will be formed, and whether corrosion in the bars of reinforced concrete will occur. More studies should be conducted to assess the potential negative impacts of chloride and sulphate ions on the long-term performance of sludge-amended cement materials, in terms of structural integrity and mechanical properties.

3.4. Supplementary cementitious material and inert addition

Generally, supplementary cementitious materials (SCMs) are classified into two categories: self-cementing and pozzolanics. Self-cementing materials react similarly to hydraulic cement, as they set and harden in the presence of water, forming solid cementitious products (Snellings et al., 2012). In comparison, pozzolanic materials (siliceous and aluminous composition) only hydrate and form cementitious compounds in the presence of moisture and an activator, usually calcium hydroxide (ASTM, 2017a).

3.4.1. Self-cementing

WTS has a potential to be used as a low-cost internal curing agent for concrete (Nowasell and Kevern, 2015). El-Didamony et al. (2014) investigated the substitution of a SCM, granulated blast furnace slag (GBFS), by alum sludge in proportions of 5%, 10% and 15% by weight for the manufacture of composite hydraulic cement (**Table 2**). The results showed that free portlandite increased in the first 7 days of curing and then decreased at 90 days. Furthermore, the replacement of GBFS by WTS led to an increase in the chemically combined water and hydration products. Compressive strength was increased by up to 5% of GBFS replacement and decreased with sludge increasing up to 15% by weight. Dahhou et al. (2018) investigated the partial replacement of PC (class CPJ55) by various amounts (5-25%) of dry Alum sludge in mortar samples with dimensions of 40 × 40 × 160 mm. They observed that the addition of 5% of WTS in PC did not affect the mineralogy of the final product. However, based on the flexural and compression tests at 28 days, mortars with 5% sludge substitution were classified as belonging to class 32.5 R according to the Moroccan Standard.

3.4.2. Pozzolanic addition

Rodríguez et al. (2010) used the atomized-dry WTS (Aluminium-based) as a pozzolanic addition in the cement mortars (**Table 2**). The mortars were prepared with the substitution of 10-30% PC by atomized sludge and tested in relation to hydration, water demand, setting time and mechanical strength. The cement replacement by the sludge considerably delayed the rates of cement paste hydration and setting time, even in the samples with only 10% of sludge. Mortars with 10-30% of atomized alum-sludge showed a substantial drop in flexural strength, with a decrease of 30% and 45% respectively. Furthermore, FTIR spectra testing showed the formation of amorphous ettringite for the montars mixes with atomized sludge. Alqam et al. (2011) investigated the use of an iron-based WTS as a partial replacement of pozzolanic Portland cement in the production of paving tiles for external use. The sludge was added in 10%, 20%, 30%, 40% and 50% (by cement weight), in making samples of 400 mm × 400 mm × 35 mm. The results showed that sludge addition did not affect water absorption of the samples, with an increase of only 6% with the maximum sludge replacement (50%). Except for samples with 50% sludge, all others obtained a minimum breaking strength of 2.8 MPa for external tiles according to the British Standard. Nevertheless, it was observed there was a decrease in mechanical strength ranging from 1.5% to 28%, when the sludge addition was increased from 10% to 50%.

Al-Tersawy and El Sergany (2016) compared the pozzolanic properties of calcined ferric-sludge and rice husk ash (RHA) in the partial substitution of cement in several concrete compositions, by ranging from 0% to 30% with two different types of aggregates (dolomite and gravel). Regarding the mechanical properties, the substitution of 10% of RHA obtained better results, with compressive and tensile strength higher than the control sample. However, the same was not observed for the replacement of calcined sludge, which obtained reductions in mechanical strength ranging from 30% to 62% after 28 days of age. The study concluded that the calcined sludge can be used in percentages less than 10% as a filler material for concrete and still reach the minimum limits for structural concrete in Egypt (ES:1524/993). In a recent and similar study, Ahmad et al. (2018) studied the feasibility of partial replacement of Portland cement by an Al-based sludge from the backwashing filtration beds, calcined at 800 °C for incorporation in cement mortar and concrete. Concerning the pozzolanic activities of the sludge, the results showed that, as well as traditional pozzolanic materials, the calcined sludge reacted with $Ca(OH)_2$ and generated significant quantities of hydrated products, and could therefore be classified as an artificial pozzolana. The addition of sludge had a small influence at the initial setting time, however, regarding the final setting time an increase of 2.6-40% was observed. The compressive strength of the mixes decreased as the sludge was added. The results showed that the replacements up to 20% could meet the Indian Standard for the paste made by Portland pozzolana cement (BIS, 2015).

3.4.3. Sand replacement

Hoppen et al. (2005) evaluated the partial replacement of sand by wet Al-based sludge in concrete mixtures. Four compositions of concrete-sludge were prepared with replacements of 3%, 5%, 7% and 10% based on the weight of the fine aggregate. The amount of mixing water in each mixing was adequate regarding the weight and moisture content in the sludge. The results indicated that 10% of sludge in the concrete is a limiting content for its practical application, first due to low workability, as well as low mechanical strength, being less than 15 MPa (**Figure 2**). The substitutions of 4-8% of sand by wet sludge in the concrete resulted in compressive strength being higher than 27 MPa at 28 days. Thus, it is suggested that the possible applications for this type of concrete should be in non-structural applications such as subfloor, blocks, non-load bearing walls, decorative concrete pieces, sidewalks, residential floors, among others. Using a similar approach, Fernandez et al. (2018) obtained results that supported Hoppen et al. (2005).

Tafarel et al. (2016) also partially replaced the natural sand present in the concrete by a wet aluminum-based sludge, in proportions of up to 10% of the dry weight of the sand. Considering the results, only the samples with 5% sludge substitution showed satisfactory compressive strength performance of 15.5 MPa at 28 days, a decrease in the strength close to 11% when compared to the reference concrete. The incorporation of 5% and 10% sludge led to an increase in water absorption by 12% and 32%, respectively. In similar research, Gomes et al. (2017) investigated the effects of aluminum-based sludge in its natural (wet) form, ranged from 0, 5, 7 and 10% of sand replacement in concrete. The results showed that the addition of wet sludge reduced mechanical strength and increased water absorption, as even 5% of sludge substitution led to a 50% reduction in compressive strength and 45% increase in water absorption.

Based on the studies presented, the substitution of natural sand for the production of structural and non-structural concretes is viable and satisfactory. The replacement of fine aggregate by up to 5% WTS in concrete and mortar led to a relatively small reduction in compressive strength (less than 20%) compared with no sludge material (**Figure 2**). Thus, 5% substitution of fine aggregate is a critical value that could be considered safe for sludge application in structural or non-structural concretes according to the Brazilian and Chinese codes (ABNT, 2014; CCES, 2005). However, further studies need to be performed to verify the influence of sludge addition on water demand, hydration of PC, and the durability of concrete.

In addition, the leaching and safety tests according to specific technical standards should be conducted to ensure the effectiveness of sludge applicability while minimizing potential damages to nature and human health.

3.5. Geopolymers

Geopolymers were designated in analogy to the raw materials (geological elements) used in their manufacture. While carbon structures form conventional polymers, geopolymers require source materials of Si and Al (Precursor), water and an alkaline reagent (activator) (**Figure A4**), which is responsible for triggering the polymerization of the components (Davidovits, 1991). Geopolymers are considered sustainable materials, mainly because both industrial wastes and by-products such as fly ash, metallurgical slag and mining residues, and geological materials such as activated clays, zeolites and kaolin can be utilized for their production (Phair and Van Deventer, 2002; Komljenović et al., 2010; Salwa et al. 2013; Singh et al. 2015; El-Eswed et al., 2017; Sudagar et al. 2018). Therefore, there have been studies on the potential of WTS as a geopolymeric feedstock, since WTS is usually rich in Si and Al oxides.

Waijarean et al. (2014) used aluminum-based WTS as alumino-siliceous material to synthesize geopolymers, aiming for their application in heavy metal waste immobilization. The results showed that the non-calcined sludge did not show compressive strength in the early ages, and after 60 days of curing achieved 0.76 MPa. The XRD profile showed that the main reason for this phenomenon was the non-occurrence of dehydroxylation of halloysite. The geopolymer samples calcined at 600 and 900 ºC presented a lower compressive strength than those calcined at 800 °C achieving 8.8 MPa after 60 days of curing.

Suksiripattanapong et al. (2015a) researched the mechanical performance of masonry units made with WTS-fly ash geopolymer, using air-dried alum sludge as aggregate, fly ash as precursor combined with alkaline activation mixture based on NaOH and Na_2SiO_3. The production of alkaline activator liquid used a fixed composition (by weight) of 9% Na_2O, 30%

SiO_2, and NaOH (10 M). The ratio of WTS/fly ash was 70:30, while the ratio of Na_2SiO_3/NaOH varied at 100:0, 90:10, 80:20, 70:30 and 50:50. Furthermore, the samples were heated at temperatures of 65, 75 and 85 °C, lasting up to 120 h and then cured at room temperature (27-30 °C). The best formulation results for maximum unit weight and compressive strength were at Na_2SiO_3/NaOH ratio of 80:20 and the alkaline activator/fly-ash ratio of 1.3. The best results were found at the temperature of 75°C and heat period of 72 h, generating samples with compression strength of 12-20 MPa. Therefore, WTS was considered feasible for the production of masonry units according to the Thai standard. Suksiripattanapong et al. (2015b) also investigated the use of the same by-products, WTS and fly ash as aggregates and precursor, respectively, for the production of a cellular geopolymer without using PC as a cementing agent. A foaming agent based on anionic surfactants, foam stabilizers and liquid air entraining were used to reduce the unit weight. The parameters used were the same as the previous study with masonry units and the influential factors studied were air content, the liquid content of alkaline activator and duration of heat and cure time. The higher compressive strengths (7.5-19 MPa) with different air contents were obtained at Na_2SiO_3/NaOH ratio of 80:20 and a heat duration of 72 h at 65 °C, which is associated with the growth of geopolymerization.

Nimwinya et al. (2016) studied the feasibility of a geopolymer precursor with calcined mixtures of WTS and RHA. Calcined aluminum sludge and RHA powder were mixed in various WTS/RHA ratios of 100:0, 85:15, 70:30, 60:40 and 50:50. The alkaline activator solution was based on the NaOH mixture with calcium silicate (Na_2SiO_3) with fixed proportions of Na_2O (8.0%) and SiO_2 (27.0%). A delay in the settling time was observed as the SiO_2/Al_2O_3 ratio was increased, mainly because of decrease in the WTS/RHA ratio. Thus, the initial settling time was increased with increasing RHA content, reaching 13.5 h for mixture with 50% RHA. The higher 7-day compressive strength (16-24 MPa) of the geopolymer paste cured at room temperature and 60 °C was reached when the ratio of SiO_2/Al_2O_3 was 4.9 and 5.9 (30% and 40% RHA replacement, respectively) (**Table 3**).

Geraldo et al. (2017) investigated the partial substitution of metakaolin (MK) by WTS for the composition of geopolymeric mortar. An alternative alkaline activator solution was prepared with NaOH and RHA (replacing sodium silicate). Furthermore, due to the amount of SiO_2 being lower in the WTS compared to the MK, an extra amount of RHA was added in the mixtures to balance and increase the SiO_2/Al_2O_3 ratio. The results showed that as the proportion of WTS was increased, setting time increased simultaneously. The final settling time of the samples ranged from 3.22 to 5.32 hours; which were higher compared to fly ash geopolymers for the same temperature. The compressive strength and the workability of the samples decreased as the addition of WTS. Mechanical strengths at all curing ages were higher than the minimum required for various types of building components, according to the Brazilian Standard (clay-fire brick > 1.5 MPa, soil-cement brick > 2.0 MPa, concrete block > 3.0 MPa).

Duxson et al. (2005) and Tang et al. (2019a) showed with aluminosilicate-base that the strength development of geopolymer matrix depended on the type of precursor and SiO_2/Al_2O_3 ratio. Thus, these few studies with WTS as a precursor have focused on improving and modifying the SiO_2/Al_2O_3 ratio when it was not satisfactory. Therefore, considering the recent research with WTS, the production of geopolymers with WTS would be a feasible solution to convert such solid waste to construction materials. However, for WTS-geopolymer to be competitive and alternative for PC-based materials, more studies need to be carried out to examine the structural performance of such geopolymers under realistic environmental conditions such as different pH, ambient temperature, humidity, alkalinity, and salt content, so as to identify potential relationships and key controls. The results obtained will provide guidance in designing further formulations such as geopolymer combinations with superplasticizers, handle retarders, polymers, natural and synthetic fibres and different alkaline activators, under different setting and curing time, so as to obtain the best products in terms of mechanical performance.

4. Durability

Natural deterioration causes decreased performance in construction materials; however, it is often considered a second or forgotten topic during the design phase (Bijen, 2003). The primary tests for the durability of materials incorporating WTS are discussed, based on studies which have been carried out on the long-term life cycle.

The durability of ceramic bricks depends primarily on water absorption (Huang et al., 2001; Hegazy et al., 2012; Ewais et al., 2017). According to Fernandes et al. (2010), the deterioration of ceramic bodies is mainly due to the high capacity of the fluid to be stored in bricks, causing the reduction of mechanical resistance and useful life. The major causes of the increased water absorption in the bricks were the burning of the organic matter present in WTS between 300-600 °C and the decomposition of carbonates and sulfates at temperatures above 800 °C, which eventually contributed to the increase in porosity (Teixeira et al., 2011; Kizinievič et al., 2013; da Silva et al., 2015). The primary test used to measure water absorption is quite similar in most of the international standards used, the majority of which limit the value of water absorption for load-bearing bricks at no more than 15-25% by weight (BIS, 1992; ABNT, 2015; ASTM, 2018). The increase in water absorption was noticeable as the sludge was added to the ceramic bodies. Among the various studies, the vast majority of which obtained water absorption values below 20%, except where the concentration of organic material in the sludge was very high (Huang et al., 2001; Teixeira et al., 2011). In addition, according to Raut et al. (2011), in their review on ceramic bricks with addition of residues and waste materials, accelerated weathering tests should also be conducted to ensure a long−term cycle for the material.

LWA and ceramsite are clay-sintered materials which exhibit similar behavior as the ceramic bricks. The water absorption, porosity and recrystallization of the liquid $CaCO_3$ are the predominant factors adversely affecting the durability of the raw materials and the light concrete produced (Huang et al., 2005; Zou et al., 2009). Huang et al. (2005) showed that among the three firing temperatures (1000, 1050 and 1100 °C) used to produce LWA made from sludge, only the specimens produced at 1100°C obtained low water absorption (14.47%), similar to commercial LWA and therefore feasible for application in concrete. Sales et al. (2010) showed that due to the high water absorption of the LWA made with WTS, all concrete formulations had relatively high porosity and water absorption (8.8%) compared to conventional concrete, which can affect the long-term life of the material depending on its exposure and application (**Figure A5**). Zou et al. (2009) demonstrated that the chemical durability of ceramsite is better when there are more complex crystalline phases and few pores. Through the tests of X-ray diffraction, morphological structures analysis, water absorption and porosity, it was confirmed that ceramsite characteristics are mostly influenced by Fe_2O_3 content, which should ideally be between 6% and 8%.

Concrete is the most commonly used construction material in the world, and its combination with steel makes it a material with broad applications. However, their durability performance depends on their interaction with the environment, where the penetration of substances through the pores has a significant impact. The most common forms of deterioration of concrete are corrosion of the steel bars through the carbonation and ingress of chlorine ions, freezing and thawing action, sulfate attack and alkali aggregate reaction (Basheer et al., 2001; Tang et al., 2019b). Therefore, the additions of sludge in the concrete must be evaluated for durability, mainly because sludge is a by-product with the presence of organic matter, chloride and sulfates ions, and other deleterious elements such as heavy metals (Cremades et al., 2018; Ooi et al., 2018). In studying the application of WTS in cement mortar, Rodríguez et al. (2010) showed, by FTIR spectroscopy test, that the presence of 12-14% organic matter (mainly fatty acids) had influenced the formation of ettringite and retardation of calcium silicate hydration process, affecting the long-term durability of concrete. In studying the addition of sludge to concrete, Hoppen (2005) reported that sludge additions above 5% caused a considerable increase in water absorption, making the material less durable and more vulnerable to penetration of chloride and sulfate ions. The concrete durability tests by accelerated aging in alkaline solution (ASTM, 2015a) with 4% of WTS showed results of potential corrosion similar to the reference concrete. However, the specimens with 8% of sludge showed a tendency of potential corrosion with 90% probability of occurrence, but no cracks or other defects were found in the concrete surface.

Concerning sulfate attack, Suksiripattanapong et al. (2015b) showed that geopolymers based on clay-fly ash had greater durability than clay-cement materials. Using the methods for wetting and drying cycles (ASTM, 2015b), Horpibulsuk et al. (2016) showed that wet-dry durability of WTS-fly ash geopolymer after 12 cycles was higher than that of the WTS-cement and silty clay–cement under the same conditions. Even after 12 wet-dry cycles, WTS-FA geopolymer with better heat curing condition (85 °C for 72 h) showed a satisfactory compressive strength of 7 MPa, compatible with durable bearing masonry blocks. Using the water absorption, voids and capillary absorption tests (ABNT, 2014), Geraldo et al. (2017) showed that the air permeability of the geopolymer with 60% substitution of metakaolin by WTS was five times higher than that specimens with no sludge addition. The porosity of the material was increased as the sludge was added, and the water absorption values were between 10.6% and 13.7%. The voids varied from 9.6% to 12%, suggesting that the sludge addition in high proportions affected the long-term behavior of the material. Up to now few studies focused on the aspect of durability and long-term effects of the addition of sludge residue. Therefore, further research is needed to assess the long-term performance of sludge-amended concrete under a range of environmental stresses such as freeze–thaw deterioration, chloride ingress, sulfate attack, and carbonation.

5. Leachability and toxicity

Considering the high potential of applying WTS in construction materials, environmental risk assessment becomes an important issue, since WTS may contain heavy metals with potential consequences for the environment and human health. The most common tests for this type of evaluation are called leaching tests, which evaluate the release of contaminants to the environment in the presence of water, in the different phases of the product life cycle (manufacturing, distribution, construction, use, end of life) and not only in aspects of 'pass fail tests' (Shih and Tang, 2011; Watanabe et al., 2011). When a leaching test is selected for material analysis, the shape of the building material tested should be taken into consideration, because the release mechanisms of heavy metals involved are different. For example, in monolithic elements, heavy metals will be released due to surface washing, diffusion and dissolution processes (NEN, 2004; Król, 2011). The most widely test applied to materials and waste is the Toxicity Characteristics Leaching Procedure (TCLP) from the United States Environmental Protection Agency (USEPA). According to Townsend et al. (2003), many chemical and physical factors control waste leaching in products. The physical factors are particle size, liquid/solid reaction, temperature and porosity. Chemical factors include pH, the redox potential of the material, sorption processes and formation of complexes with organic or inorganic compounds. Considering the large amount of literature evaluated in this review, only limited studies have assessed the environmental risk caused by the incorporation of various types of WTS into building materials (**Table 4**). This may be due to the fact that in most cases the sludge is classified as non-hazardous waste according to local standards (Hidalgo et al., 2017).

In a study with arsenic-iron sludge for the production of bricks, Hassan et al. (2014) reported that for different pH only sludge-mix ratio of 3% showed less arsenic and iron leaching than was acceptable by the Bangladesh and World Health Organization (WHO) Standards. The highest values for leaching of the burned brick were found at pH around 3.0, showing values above the limit values of 0.05 mg/l for arsenic. Sales et al. (2010; 2011) studied the leaching of aluminum, iron and lead in a LWA produced with Al-based WTS and sawdust. The solubilized extract of the concrete produced with the WTS-LWA showed an aluminum concentration (19.96 mg/l) which is substantially higher than the solubilized extract concentration of the reference concrete without this composite (1.12 mg/l). The result was mainly due to the high concentration of aluminum (11.10 mg/l) present in the sludge. Nevertheless, the material showed the necessary limits for environmental safety according to the NBR 10004 Brazilian standard (ABNT, 2004) and classified as non-harmful and non-inert solid wastes. Xu et al. (2008c; 2010) studied the stabilization of heavy metals in ceramists made with Al-WTS and WWTS. Leaching tests focused on the effects of pH, oxidative condition, sintering temperature and the ratio of oxides (Fe_2O_3 + CaO + MgO)/(SiO_2 + Al_2O_3). The effect of sintering temperature and the ratio of oxides had a great influence on the leaching and solidification, noting that above 1000 °C heavy metals such as Cd, Cr, Cu, and Pb can hardly be re-released to the environment causing secondary pollution. The results also show that sample-ceramsites exposed to conditions of pH > 2 had leaching rates below those recommended in the Chinese Standard and TCLP. Similar findings were reported by Wei (2015) that the heavy metal solidifying efficiencies of sludge-derived LWA were strongly enhanced by crystallization and chemical incorporations within the aluminosilicate or silicate frameworks during the sintering process. Therefore, it is concluded that heavy metals can be properly stabilized in LWA samples containing sludge and prevented from release into the environment again to cause secondary pollution.

When studying eco-cements clinkers made with two industrial waste and Al-based WTS ash, Lin and Lin (2005) analyzed the concentrations and leachings of heavy metals Pb, Zn, Cd, Ni, Cr and Cu. The results confirm a high stabilization rate of heavy metals, and all eco-cement mixtures were below the regulatory limits established by TCLP. Similar analysis was made by Chen et al. (2010) in their study of shale replacement by WTS for clinker production. The TCLP results showed that for the hydrated samples made with the WTS-clinker, no heavy metal was detected above the normalized standard, indicating that these components were fixed and incorporated to the clinker. Lee et al. (2012) analyzed the leaching effects of the substitution of natural sand by an Al-based WTS in the production of concretes, and the results of TCLP analysis showed that no substantial quantities of heavy matter were detected in the concrete samples. Proving that, even when large quantities of sludge (30%) were added, the heavy metals were fixed and immobilized in the concrete matrix. Using TCLP, Alqam et al. (2011) evaluated the heavy metals leaching of paving tiles made from the mixture of Al-sludge and cement. All results showed very low percentage of heavy metal concentrations, even for paving tiles with 50% Al-WTS contents.

Waijarean et al. (2017) evaluated the leaching of Zn, Fe and Cr in geopolymers synthesized by alkali activation of WTS. Leaching tests in simulated acid rain indicates that the geopolymers with molar ratios of SiO_2/Al_2O_3 = 1.78 were capable of immobilizing all three metals to within safe leachate levels in accordance with USEPA regulatory limits. It was observed that as the SiO_2/Al_2O_3 ratio was increased, the WTS-based geopolymer became less effective as an immobilizing agent. For Cr (the most toxic of the 3 metals), the results simulating acid rain (pH 5) showed that at SiO_2/Al_2O_3 ratio of 3 Cr concentration in the leachate reached 8.9 mg/l, exceeding the USEPA recommended value of 5 mg/l.

6. Economic evaluation

In their research on WTS application in structural ceramics, Wolff et al. (2015) showed that for every ton of clay, it would be possible to save US$318 by introducing sludge into the brick production cycle. Included in this amount is a saving of US$8/ton that would be spent in transport to the landfill and other cost associated with clay extraction and energy. Kaosol (2010) evaluated the cost involved in the production of hollow concrete blocks with fine aggregate replacement up to 50% by using WTS in Thailand. The cost composition for the evaluation was based on the production of 1200 blocks day, 2 laborers, machine cost, electricity and materials for normal block production (cement, sand and aggregate). In their evaluation, for the replacement of 10% and 20% of fine aggregate by WTS, the cost per unit would be reduced by Thai-Baht ฿0.64 and ฿1.05 respectively, for a hollow load bearing concrete block. Furthermore, for the manufacture of non-load bearing blocks with 30-50% of sludge, the total costs would be reduced by Thai baht ฿1.48-2.35 per block.

Lima and Zulanas (2016) performed an economic feasibility analysis for the incorporation of 5% WTS in the concrete mix sidewalks in Cubatão city, Brazil. The analysis considered the Cubatão city's dry-sludge production of 1925 kg/day and a standard city block of 80 m × 274 m (1534.4 m³ for sidewalk of 7 cm thickness). According to their analysis, it would need three days of sludge production to supply the required 5% addition in the 98-m^3 concrete mix for 700 m of a sidewalk, and 39 days of water filtration process to achieve enough WTS to lay a city block of concrete. Thus, in 1 year with a production of 9.4 city blocks of sidewalks (365 days in a year/39 days to produce sufficient sludge for a city block), the city could consume all its sludge production of the year. In addition, according to the researchers, the local civil construction industry could save $543069/year with the assumption that 1 ton of concrete gravel (washed rock) is $28.81 with 5% WTS substitution. Gastaldini et al. (2015) analysed the costs (US$56.04/m^3) of collecting/transporting the sludge from water purification plant to the landfill, and the fees applied to allocate the sludge in the Rio Grande do Sul, Brazil. They estimated that the cost to carry 1 kg of sludge to the landfill was twice as the cost of a 19-mm coarse aggregate (US$0.018) and 5.6 times the cost of washed and sieved sand (US$0.0083). Therefore, the 5-30% substitution of cement by WTS ash (for the same concrete strength without the addition of sludge) would lead to savings of 37-200 kg of cement per m^3 of concrete.

In evaluating the production of geopolymer binders with WTS, Nimwinya et al. (2016) showed that the cost involved would be twice as practiced today in Thailand which is US$33/ton, when WTS is transported to landfill sites (although actual landfill cost is not included). This relatively high cost is mainly related with the geopolymers ingredients, calcination of the sludge and the electricity used in the curing process, which stands at US$65 and US$68 for geopolymers prepared at room temperature and cured at 60 °C, respectively. However, such analyses have not considered the significant savings in CO_2 reduction, when

WTS is replacing natural raw materials in construction material production. Furthermore, more financial gains can be obtained by the recovery of metals e.g. Al from WTS (Ooi et al., 2018), and the sales of geopolymers (Nimwinya et al., 2016).

7. Conclusions

This paper critically reviewed the research progress in the application of WTS in construction materials. Some conclusions can be drawn up as follows:

- The latest research results on the application of WTS in a range of construction materials were critically examined. In summary, WTS has some unique characteristics to enable its incorporation in construction materials such as bricks, ceramics, LWA, cement and geopolymers.
- The main challenges in the application of WTS in construction materials are derived from its high variation in physicochemical properties and relatively high content of organic matter, which will increase the porosity and water absorption therefore potentially adversely affecting products' structural integrity and performance.
- The majority of studies with ceramic bricks showed that the incorporation of alum sludge up to 10% is satisfactory in maintaining the structure performance. For iron-based sludge, an increase in the mechanical strength was observed when more significant proportions were added. A similar effect was observed in studies with LWA, where Fe content in the sludge had a high impact on water absorption and compressive strength.
- Regarding the production of cement and concrete with the partial addition of WTS, it has been shown to be potentially feasible, especially as a pozzolanic additions and replacement of sand in concrete up to 5%.
- The effect of sintering temperature and the ratio of oxides had a great influence on heavy metals leaching from WTS-amended products. Sintering above 1000 °C is recommended

to ensure the full immobilisation of heavy metals such as Cd, Cr, Cu and Pb hence preventing further environmental pollution.

- Used as a substitution, WTS can make saving for raw materials and energy, replace landfill as solid waste management, and contribute to sustainable construction materials production.

8. Recommendations for future research

Extensive research has been conducted with satisfactory results on the application of WTS in construction materials. However, due to the complexity of this material, much remains to be done to improve its incorporation in the construction industry practice. As far as the sintered material (Bricks and LWA) is concerned, the best results were achieved with WTS blended with materials rich in silica (RHA, glass). Therefore, with more conclusive studies and commercial scale applications of such combinations, the practice could provide a solution to the sound management and valorisation of WTS without adverse effects on the mechanical performance of modified construction materials.

Most of the studies have examined the engineering performance of WTS-incorporated cement and concrete in terms of mechanical strength, setting time, chemical-mineralogical characterization and leaching tests, in a relatively short time window. There is a lack of long-term durability testing in concrete, which should be addressed in future studies. In addition, due to the presence of certain substances in sludge such as chloride and sulfate ions which are potentially deleterious to concrete, there is no guarantee that the use of WTS will not develop expansive products (Ettringite) or corrosion in the bars of reinforced concrete. Further studies on this topic should be carried out to ensure the safety and sustainable use of sludge residue.

Considering the fact that research on WTS-derived geopolymers is a relatively new approach, future studies should explore ways to improve their mechanical strength and to resolve the problems associated with setting time and workability. More studies are needed to explore appropriate experimental conditions such as combination with superplasticizers, polymers, fibres and different alkaline activators (KOH and K_2SiO_3). Furthermore, potential leaching of toxic chemicals and material durability from alkali-silica reaction, efflorescence, acid/sulfate attack, carbonation and steel reinforcement corrosion need to be addressed urgently, to promote the sustainable recycling of WTS and solid waste in general in construction materials.

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