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Valorization of sewage sludge in the fabrication of construction and building materials: a review Zhiyang Chang^a, Guangcheng Long^{a*}, John L. Zhou^{a,b*}, Cong Ma^a ^aSchool of Civil Engineering, Central South University, 68 South Shaoshan Road, Changsha, Hunan 410075, China ^bCentre for Green Technology, School of Civil and Environmental Engineering, University of Technology Sydney, Sydney, NSW 2007, Australia

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Abstract

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With increasing amount of sewage sludge becoming an urgent and inevitable issue for every country, its applications in the production of construction and building materials provide an alternative solution for sludge disposal and resource recovery. Similar to clay and Portland cement, the main oxides in sewage sludge are SiO₂ (10-25%), Al₂O₃ (5-10%) and CaO (10-30%) which are increased in sludge ash after incineration to 25-50%, 10-20% and 15-30%. Therefore, this solid waste can be utilized not only as raw material for the production of eco-cement, bricks, ceramic materials and lightweight aggregates through sintering process, but also as supplementary admixtures in cementitious materials such as pozzolanic component, fine aggregate or filling material. By critically reviewing current utilizations of sewage sludge, it is feasible to replace up to 15% natural raw materials with sewage sludge in cement production and the manufactured eco-cement clinkers show comparable performance to traditional Portland cement. Whilst as raw feed in the fabrication of bricks, ceramic materials and lightweight aggregates, 20% of sewage sludge substitution is acceptable to produce good quality products (within 8% firing shrinkage and 15% water absorption). Though high content of organic matter in raw sludge causes a decrease in mechanical strength and delay in hydration process, controlled low-strength materials offer an innovative reuse with large amount of sludge. The immobilization of heavy metals in products prevents sewage sludge causing secondary environmental pollution. Furthermore, suggestions for future research are proposed in order to strengthen the high value-added applications of sewage sludge.

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Keywords: Sewage sludge; Construction materials; Mechanical properties; Leachability; Durability

1. Introduction

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The rapid growth of world population, economic development, industrialization and urbanization necessities sewage treatment through primary sedimentation, secondary treatment, and increasingly tertiary treatment (Christodoulou and Stamatelatou, 2016; Yang et al., 2015; Zhou et al., 2009). During sewage treatment process, large quantities of sewage sludge are produced worldwide (Fig. 1). In the EU, the highest annual sewage sludge production with 1.85 million tons of dry solids was in Germany, followed by the UK with 1.14 million tons and Spain with 1.03 million tons (Kelessidis and Stasinakis, 2012). In comparison, more sludge is produced in Japan than in EU countries. With limited data from the US, its sludge production is ranked number two. With the world's biggest population (1.3 billion), the production of sewage sludge in China has increased continually since 2011, and reached over 12 million tons of dry sludge solids per year in 2017 (MHURD, 2017). In 2017, there were 45 million tons of dry solid sludge output all over the world (Zhang et al., 2017). Thus, the treatment and disposal of sewage sludge is becoming an urgent issue that the world is facing today. Sewage sludge from sewage treatment plants (STPs) is highly heterogeneous in composition, and typically contains many organic and inorganic substances including microbial biomass, pathogens, N and P nutrients, metals and sediments (Zaker et al., 2019). In addition to a high water content up to 95%-99% (Yang et al., 2015), high concentrations of heavy metals such as Pb, Cd, Cr, Hg and Ni are detected in sewage sludge (Belhaj et al., 2016; Mulchandani and Westerhoff, 2016). The environment and human healthy would be adversely affected if the sewage sludge is discharged directly into surrounding environment without any treatment. Therefore, the treatment of sewage sludge is indispensable before its disposal, to ensure the volume reduction, stability and valorization of sludge. Currently, the main processes of sludge treatment include sludge thickening, conditioning,

dehydration, stabilization and drying, with a variety of physical, chemical and biological methods (Liu et al., 2013). Generally, the thickening process can remove the pore water in sludge and reduce the moisture content to about 95%. The purpose of conditioning is to improve the sedimentation and dehydration properties of sludge by changing its microstructure. Although the pore water can be removed by thickening, the volume of sludge is still large. In order to reduce the moisture content and volume of sludge further, dehydration process will be carried out by natural drying and mechanical dewatering. Moreover, the putrefaction of sewage sludge is another problem for producing foul smell and breeding bacterium as a result of the high organic matter content in sludge. Stabilization treatment containing aerobic and anaerobic digestion, compost, alkaline treating and high-temperature pyrolysis can decompose organic matter by biological and chemical processes. Sludge drying technology can realize the considerable reduction of sludge volume, improve the heat value of sludge, and reduce the harmful components such as microorganisms and pathogens, therefore creating conditions for transportation and incineration or resource utilization of sludge.

There are several approaches for the recovery or disposal of sewage sludge after the treatment, including incineration, landfill, agricultural application, compost and other applications (Tyagi and Lo, 2013; Belhaj et al., 2016). Fig. 2 presents the status of sludge disposal in selected countries. In Australia, the US, China, Norway, France, Spain and the UK, agricultural use of sewage sludge is the dominating route. In Germany, Poland and Austria, the application of sewage sludge in agriculture is limited by their current regulations with high standard of sludge quality, hence sludge is mainly disposed by modern incineration and other methods (Mininni et al., 2015). In Japan, the amount of sewage sludge used in the manufacture of construction materials accounts for 48% of the total sludge production. However, improper disposal of sludge such as dumping at sea or land directly, is still in existence in some countries (Yang et al., 2015), which would cause serious secondary pollution.

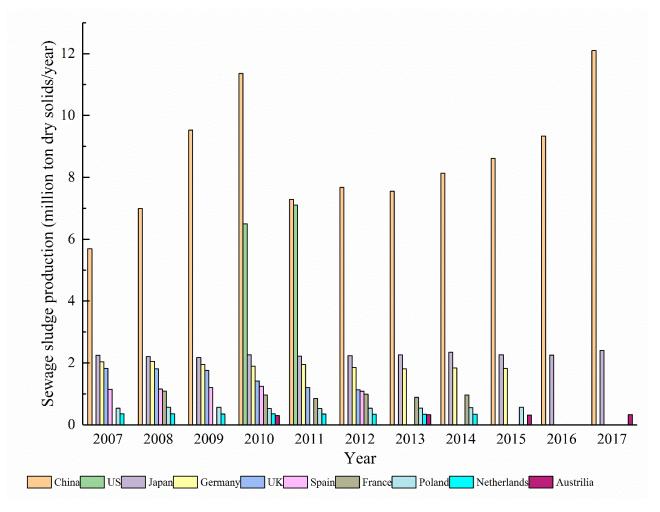


Fig. 1. Estimated yearly productions of dry sewage sludge from selected countries. Data from Eurostat Statistics (2015), MHURD (2017), MLRT (2017), EPA (2018), NSBPEU (2016).

Sewage sludge has a similar mineralogical composition to clay and Portland cement as it contains major oxides such as SiO₂, Al₂O₃, CaO and Fe₂O₃. Based on chemical composition, sewage sludge is used widely in the production of construction and building materials such as eco-cement, bricks, ceramic material and lightweight aggregates (LWAs) or supplementary cementitious materials SCMs) (Gherghel et al., 2019; Lynn et al., 2015; Świerczek et al., 2018). These applications offer alternative methods for the recycling of sludge and resource saving in the long term. However, the amounts of sewage sludge used in current applications are only a small proportion of total sludge production, and sludge-based productions are generally of lower quality and cause environmental

concerns such as heavy metal leaching. Many review articles and books have been published on sewage sludge waste (Fytili and Zabaniotou, 2008; Smith et al., 2009; Donatello and Cheeseman, 2013; Lynn et al, 2015; Smol et al., 2015; Świerczek et al, 2018; Dhir et al., 2017). However, to our knowledge, so far there is no article specifically reviewing the utilization of both raw sewage sludge and incinerated sludge ash in building materials based on their characteristics. Therefore, this review article aims to critically evaluate the latest development, trends and challenges of applying this waste material in building and construction materials such as cement, concrete, ceramic bricks, LWAs and SCMs. In addition, this review assesses the durability and environmental risks of sludge-amended products specifically the leachability and toxicity of heavy metals. Finally, suggestions for sustainable and high value-added utilization of sewage sludge in future research are also discussed.

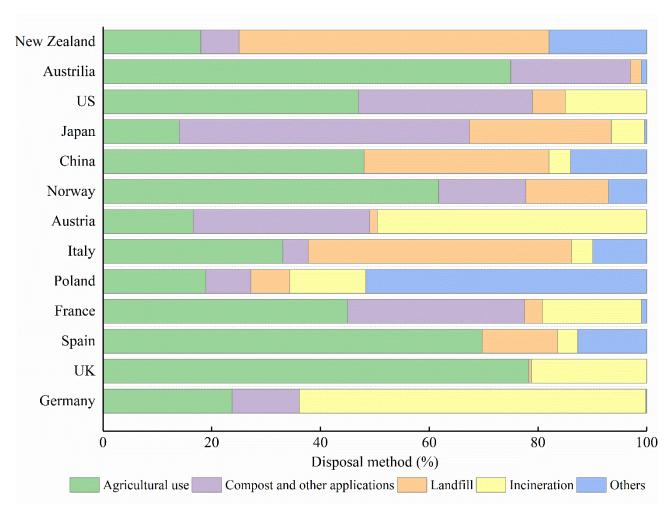


Fig. 2. The implemented disposal methods of sewage sludge from selected countries, compiled from Eurostat Statistics (2015), MHURD (2017), MLRT (2017), EPA (2018), NSBPEU (2016).

2. Characteristics of sewage sludge

Sewage sludge is produced from primary settling tank and secondary sedimentation tank in STPs, consisting of a variety of organic and inorganic substances including excess microbial biomass (Cieślik et al., 2015; Kulikowska and Gusiatin, 2015). Its composition is highly complex as a result of various input sources, and treatment technology adopted. Sewage sludge is a liquid or semi-liquid waste with high water content (55-80% for dehydrated sludge) and the content of organic matter usually accounts for 60-80% in dry solids of sewage sludge resulting in high loss on ignition. In addition, inorganic and organic contaminants such as heavy metals (Belhaj et al., 2016) and endocrine disrupting chemicals (Zhou et al., 2009) are often present in sludge, the content of which mainly depends on the amount of industrial wastewater discharged into STPs. However, a study by Xu et al. (2014) indicated that the presence of phosphorus and other trace elements provided favorable condition for the formation of tricalcium silicate (C₃S) in cement clinker by increasing the amount of liquid phase and decreasing the viscosity.

For the application of sewage sludge in building materials, the mineral composition and geotechnical property of this waste becomes a key point in numerous studies. The major oxide components of sewage sludge are SiO₂, CaO, Al₂O₃, Fe₂O₃, MgO and P₂O₅, although its precise composition and quality may vary significantly depending on the source of the sewage and the types and dosages of additives introduced into sludge treatment process. Fig. 3 presents a ternary diagram of the main oxide contents of sewage sludge and incinerated sewage sludge ash samples from various studies along with typical contents for well-established cementitious materials. Compared to raw

sewage sludge, sewage sludge ash has higher content of SiO₂, CaO and Al₂O₃ which are comparable to latent hydraulic materials (e.g. granulated blast furnace slag), pozzolanic materials (e.g. pulverised fuel ash) and filler materials (e.g. ground limestone) as cementitious components (Dhir et al., 2017a). In addition, a high content of Fe₂O₃ can be observed in sewage sludge which is highly favorable for the production of cement, bricks and ceramic materials, as a result of saving iron ore when using sewage sludge as a raw construction material (Montero et al., 2009; Qi et al., 2010; Zhang et al., 2016). Lime (CaO) is used widely for the dewatering and drying process of sewage sludge resulting in a high content of CaO in sludge which could potentially be used as an alternative for limestone in cement production (Xu et al., 2014). However, the high content of organic matter in sewage sludge may affect the cementitious property leading to high porosity and low bonding strength, so pretreatment of raw sludge is often conducted before being used in cement or concrete. For example, thermal treatment and incineration are studied frequently for more effective utilization of sewage sludge in building materials. Furthermore, the solidification of heavy metals can be also obtained at high treatment temperatures in firing process.

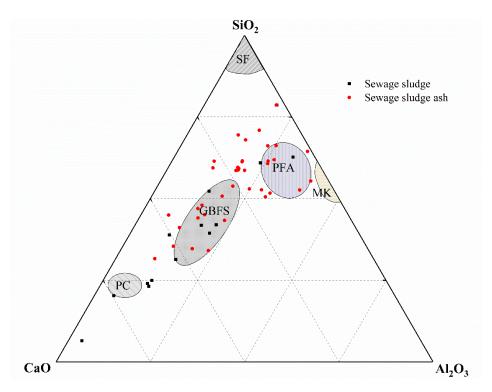


Fig. 3. Ternary plot of SiO_2 , Al_2O_3 and CaO contents for sewage sludge and incinerated sludge ash. PC = Portland cement, GBFS = granulated blast furnace slag, PFA = pulverised fuel ash, MK = metakaolin, SF = silica fume.

3. Application of sewage sludge in construction materials

3.1. Cement clinker production

As cement is the most widely used building material in the world, cement industry is often regarded as having excessive energy consumption and serious environmental pollution. It is true that cement plants have become the main contributors of energy and natural resource consumption as well as CO₂ emissions, especially in developing countries (Aprianti et al., 2015; Oh et al., 2014; Shi et al., 2011). Currently, there is an extensive research interest in eco-cement as alternative SCMs manufactured with waste materials such as municipal solid waste (Garcia-Lodeiro et al., 2016; Lin et al., 2003), construction and demolition waste (Mymrin and Corrêa, 2007) and industrial by-products (García-Lodeiro et al., 2013; Part et al., 2015) in order to reduce their environmental impact. Sewage sludge is used in cement production process or as a SCM due to its similar mineral and chemical

components to Portland cement (Tay and Show, 1997; Zabaniotou and Theofilou, 2008). In Japan, the amount of sludge for Portland cement production was about 20% of total dried sewage sludge yield in 2017, according to MLRT (2017).

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Tay and Show (1991) studied the feasibility of using dewatered sewage sludge mixed with lime to produce a cement-like material through the incineration at 1000 °C for 4 h in a furnace. They found that compressive strengths of the mixtures with sludge-to-lime mix proportion of 1:1 could satisfy the requirements of the ASTM standard for masonry cement. Table 1 represents the main sintering parameters and properties of eco-cement in various studies conducted on sewage sludge utilization in cement clinker production. Rezaee et al. (2019) investigated the physical, chemical and mechanical characteristics of eco-cement produced by dry municipal sewage sludge as partial substitute from 5% to 15% of traditional raw materials. The major chemical components of ecocements were similar to ordinary Portland cement but the amount of water demand and initial and final setting times were increased. Furthermore, Xu et al. (2014) used lime-dried sludge as a substitute of limestone material in cement production sintering at 1400 °C. They reported that the introduced trace elements in mixing sludge played an important role as mineralizers and cosolvents in cement sintering by reducing the eutectic point of the system and accelerating the formation of liquid, which was beneficial for forming tricalcium silicate (C₃S). However, these improvement effects would be weakened significantly if the amount of lime-dried sludge added reached up to 18 wt% and the formation of main crystalline phases in cement clinker would also be hindered because of the excessive amount of sludge (Lin et al., 2009). Shih et al. (2005) also found that the use of heavy metal-containing sludge as the replacement of raw material for cement production was feasible, since the introduced heavy metals could enhance the formation of C₃S phase in cement as the addition of sludge was within 15% which would not cause a leaching risk from the sintered clinkers. Therefore,

to ensure the strength requirements and environmental safety, the amount of sewage sludge added as raw materials in cement clinker production should be stringently controlled within a certain range (≤15 wt%). On an industrial scale, the co-processing of sludge in cement kiln has been considered as a sustainable way to dispose sludge in China, and 65000 tons of sludge had been successfully used in a new cement plant during 2008-2012 (Li et al., 2012).

As an alternative replacement of clay, sewage sludge is also combined with other solid waste in cement production. Lin et al. (2004; 2005) used different types of waste sludge ash, including sewage sludge ash, water purification sludge ash and steel slag and limestone, as raw components for the production of eco-cement clinkers by burning at 1400 °C for 6 h. The major components of ordinary Portland cement, C₃S, C₂S, C₃A, and C₄AF, were all found in new clinkers. The results indicated that it was feasible to use sludge ash to replace up to 20% of raw mineral components. Similarly, a mixture of various waste sludge including marble sludge, sewage sludge, drinking water treatment plant sludge, and basic oxygen furnace sludge was used as the raw materials for the production of eco-cement (Yen et al., 2011). The addition of marble sludge provided a sufficient amount of CaO to form more C₃S crystalline phases, which contributed to early strength development of cement paste.

Sewage sludge can be used not only as raw material but also as alternative fuel in clinker production due to its high calorific value (Zabaniotou and Theofilou, 2008), which is especially the case for dried sludge with high content of organic matter. Valderrama et al. (2013) evaluated the environmental impacts of the sewage sludge as alternative fuel and raw material for cement production by life cycle assessment methodology. The results showed that CO₂ emission was reduced when the amount of fuel substitution was increased from 5% to 15%. Except for CO₂, a reduction of NO_x emission was also investigated when co-processing sewage sludge in cement kiln, but the

emission of total semi-volatile organic compounds increased with the increasing amount of sewage sludge (Lv et al., 2016). Another study (Fang et al., 2015) also indicated that it was conducive to NO_x reduction with the application of sewage sludge as a denitrification agent and secondary fuel in cement production. Though resource conservation is realized when using sewage sludge as an alternative fuel in cement plants, the environmental risk assessment and evaluation of potential human health risks also deserve more attention. Several studies (Hong and Li, 2011; Nadal et al., 2009; Rovira et al., 2011; Schuhmacher et al., 2009) indicated that the utilization of sewage sludge as a combustible material represented an environmental improvement without additional health risk for human, and such a practice was acceptable according to international standards.

Based on reports from various studies, the reuse of sewage sludge as raw material for cement clinker production is feasible. This is because that the main minerals in sludge such as calcite and kaolinite provide sufficient amount of feed to form crystalline phases of cement, and the introduced trace elements of sludge enhance the formation of cement clinker components. Furthermore, the dried sludge with a high organic matter content can act as a fuel due to its high calorific value. However, the excessive addition of sewage sludge decreases compressive strength and increases water demand and final setting time of eco-cement. It is therefore recommended to use a maximum sludge addition of 15% as a safe limit, so that eco-cement clinkers comprise similar chemical composition to Portland cement and present good workability and comparable mechanical properties to traditional cement paste. This review also identified the sintering temperature and duration of eco-cement as another important parameter, for which, currently, there are no uniform standards or rules. low temperature may result in low pozzolanic activity of sludge while high temperature causes excessive energy use. Furthermore, the long-term mechanical performance of sludge-substituted cementitious materials and

- the potential risk of heavy metals leaching are not extensively studied, hence a review is timely to
- summarize the progress and challenges in these aspects.

Table 1. Summary of main sintering parameters and properties of eco-cement incorporating sewage sludge

Raw material	Sludge content (%)	Calcination		Main property			
		Temperature (°C)	Time (min)	Compressive strength (MPa)	Flexural strength (MPa)	Final setting time (min)	Reference
Sewage sludge, limestone, marl, bauxite, iron ore	15	1400-1500	20	60.5	8.4	185	Rezaee et al., 2019
Sewage sludge, iron, shale, limestone, fly ash, sand	15	1450	120	60.48	9.55	263	Lin et al., 2012
Lime-dried sludge, iron slag limestone, clay	15	1400	60	61.8			Xu et al., 2014
Sewage sludge ash, water purification sludge ash, limestone, ferrate	16.7	1400	240	62.5			Lin et al., 2005
Marble sludge, sewage sludge, drinking water sludge, basic oxygen furnace sludge, sand, limestone, clay, iron slag	30	1000-1400	60	67.5			Yen et al., 2011
Heavy metal-contaminated sludge, surface finishing sludge, electroplating sludge, clay, limestone, ferrate	15	1400	180				Shih et al., 2005

3.2. Supplementary cementitious materials

SCMs are commonly used in concrete mixtures as a partial replacement of clinker in cement or as cement in concrete. Generally, the function of SCMs is achieved by two approaches: self-cementing and pozzolanic reaction (Gomes et al., 2019). These two effects can be observed when sewage sludge or sewage sludge ash is used as a replacement of cementitious materials in concrete. Although sewage sludge ash has lower organic matter content and higher pozzolanic activity compared to raw sewage sludge, it requires additional thermal treatment which inevitably involves additional energy consumption and cost. Therefore, the applications of raw sewage sludge in construction industry are explored first in this part.

Sewage sludge

As for the utilization of sewage sludge with Portland cement, the property of a matrix with sludge and cement was evaluated by Valls and Vazquez (2000). Portland cement was partially substituted by variable amount of sewage sludge from 25% to 50%. The results showed that there was no obvious difference in the hydration products between different pastes with and without sewage sludge except for the presence of hydrated calcium carboaluminate in the sludge samples. The setting process of the system was significantly hindered by high organic matter content in sludge so that the beginning and end setting time were delayed from 2 h and 3 h to 15 h and 23 h, respectively. Furthermore, the alkalinity of the cement system promoted the decomposition of organic matter contained in sewage sludge from a long-term observation. In view of the obvious setting retarding effect, calcium chloride (CaCl₂) and calcium hydroxide (Ca(OH)₂) were also added into the mixes of sludge and cement as accelerating additives in the study by Malliou et al. (2007). The addition of CaCl₂ in the mixed paste shortened the setting time significantly and therefore improved the early compressive strength with the optimum mixing amount of additives (3% CaCl₂ and 2% Ca(OH)₂ of cement weight). In addition, Pavšič et al. (2014) used the biomass ash for the solidification of raw sewage sludge with a low content of dry solids to produce a low strength material with a compressive strength of 1.8 MPa.

Moreover, recycled aggregates were also added in the composites to produce a similar low-strength material which could be used in applications such as landfill cover and road foundation.

In the work by Rahman et al. (2017), the sludge collected from the local textile industries was utilized as an alternative material for cement or fine aggregates to produce mortar and concrete. Their results showed that the mortar specimens with 10% replacement of cement or 50% replacement of sand by textile sludge separately showed 50% or 45% reduction in compressive strength, compared to mortar without sludge. In another study by Balasubramanian et al. (2006), the results also indicated that textile sludge could be used in mortars and concretes for non-structural building components where lower strength was allowed and the substitution of textile sludge for cement could reach up to a maximum of 30%. Furthermore, the feasibility of increasing the proportion of textile sludge up to 35% mixed in concrete to make the non-structural building materials was verified by experimental study (Garg et al., 2014). The utilization of sludge in asphalt mixture was also investigated in several studies (Akbulut et al., 2012; Zhang et al., 2012). Lucena et al. (2014) added sewage sludge into soil with addition of different additives such as lime, cement and bitumen to make a modified soil which was used for base of road pavements. Their tests indicated that the mix containing 1% of bitumen presented the best compressive strength due to the fact that bitumen used as a binder increased grains cohesion.

In recent years, many researchers have explored the potential utilization of various solid wastes, for example coal bottom ash (Katz and Kovler, 2004), coal fly ash (Lee et al., 2013) and rice husk ash (Nataraja and Nalanda, 2008) in the production of controlled low-strength materials (CLSM) which are widely used in different kinds of construction structures (Horiguchi et al., 2010; Siddique, 2009). Sewage sludge could also be used to make CLSM after the condition, stabilization and solidification processes. Traditionally, fly ash, lime and cement are widely used as solidifying agents to attain high strength mixtures with sludge (Lim et al., 2002; Yin, 2001). Kim et al. (2005) applied converter slag and quick lime in sewage sludge and made solidified sludge used for landfill cover material. Their results showed that hydrated calcium silicate (CaO·SiO₂·nH₂O) was the main

hydrated product in mixtures and some harmful bacteria could be eliminated by the solidification process. Li et al. (2014) studied the geotechnical properties of the dewatered sewage sludge conditioned with skeleton builders, such as fly ash and lime combined with ferric chloride, which was reused as landfill cover materials. The application of fly ash and lime not only improved geotechnical properties of sludge with high plasticity index and low permeability coefficient but also accelerated the formation of hydrated products which contributed to the mechanical property such as compressive and shear strength. Furthermore, in another study by Hwang et al. (2017), the alkaliactivated CLSM was produced by using fly ash, ground GBFS and sewage sludge with sodium hydroxide (NaOH) as activator. The addition of sewage sludge resulted in increasing fresh unit weight and compressive strength of the CLSM samples while it reduced the workability as well. Additionally, the alkali equivalent played a significant role in the setting times of the CLSM which was also observed by Lee et al. (2013).

In consideration of the high moisture content of sewage sludge, it is observed that using it as a replacement of SCMs can not only provide the water demand for the production of cement mortars and concretes but also eliminate the prior process of dewatering and drying of raw sludge (Hamood et al., 2017). The feasibility of using raw sludge as a water replacement in cement-based materials was investigated by Hamood et al. (2017) who collected raw sewage sludge from a STP containing 97.5% liquid, which was mixed with unprocessed fly ash with high carbon content and large particle size to replace cement with various proportions. Contrast with the mortar mixtures with water, the flowability and total water absorption of the mixtures with sewage sludge was comparatively lower, while the compressive strength decreased noticeably by 40%.

Generally, raw sewage sludge has low pozzolanic activity and the existence of organic matter causes a delayed formation of main hydrated products, resulting in negative effect on the setting and mechanical properties of cement-based mixtures. Therefore, sewage sludge material without any pretreatment may not be suitable to be used directly as additive material or fine aggregates in cement for good performance. However, it is identified from the review that CLSMs offer an innovative reuse

with large amount of sewage sludge where low strength of the structure is allowed. The liquid sludge with high content of moisture could provide the essential water for the production of building materials by conditioning the quality of sewage sludge. In addition, the potential geotechnical property of sewage sludge could be improved by combining with other pozzolanic materials, an area which offers exciting research opportunities.

Sewage sludge ash

As concluded from the aforementioned work, the presence of diverse organic matter in sewage sludge poses a threat to the mechanical properties such as the setting time, compressive strength and durability of sludge-modified construction materials. Furthermore, most mineral compositions in raw sludge are in low activity and make few contribute to strength development of cement or concrete. Therefore, the thermal treatment (mostly referring to incineration) of raw sludge is widely used to obtain sludge ash before the mixing procedure. During firing or incineration, the organic matter is decomposed, and inert minerals such as kaolinite are activated by being converted to metakaolin at high temperature. Thermal treatment aims to alleviate adverse effects on the mechanical strengths of construction products and immobilize potentially the heavy metals in sludge. In Europe, the sludge incineration is the most commonly used method of sludge treatment while a large amount of sludge incineration ash will be produced during the process (Fytili and Zabaniotou, 2008).

Cyr et al. (2007) investigated the physical, chemical and mineralogical characteristics of sewage sludge ash and evaluated the feasibility of its use in cement-based materials. Results showed that the high specific surface area of sludge ash particles increased water demand significantly and the early cement hydration was delayed by minor elements in the ash, resulting in decreasing early compressive strengths of mortars. However, the potential pozzolanic content in sludge ash contributed to long-term development of strength. Other than the reactions of other by-product materials with Portland cement, the amorphous or poorly crystalline hydroxyapatite (Ca₅(PO₄)₃OH) was formed in the mixture with sewage sludge ash except for large quantities of Al₂O₃-Fe₂O₃-mono (AF_m) phases in the work of Dyer et al. (2010). Chen and Poon (2017) compared the properties of cement mortars blended

with sludge ash and fly ash separately. They observed that sewage sludge ash with smaller particles and higher specific surface area accelerated the cement hydration as it provided more nucleation spaces for hydration product precipitation. The workability of mortar decreased as a result of the hydroscopic characteristic of sludge ash particles caused by the porous nature while the ball-bearing effect of fly ash increased workability (Zheng et al., 2016). Furthermore, the compressive strength of mortar with 20% sludge ash at 90 days merely decreased by 4.5% comparing to the control mortar without sewage. Both the pozzolanic activity and water retention effect of sludge ash particles resulted in this result. Sewage sludge ash particles could absorb water into its pores at early age and release the water gradually from pores later (Chen and Poon, 2017). Apart from similar hydrated product components to cement, brushite generated from the reaction of amorphous iron phosphate and calcium hydroxide was found in the mixture on account of high Fe₂O₃ and P₂O₅ content in sludge ash (Sopcak et al., 2016). The brushite, also called whitlockite (Ca₃(PO4)₂), was found in the hydrated products of mixtures with sludge ash which contributed to the development strength of mortar as well (Donatello et al., 2010). Fig. 4 summarized the effect of incinerated sludge ash used as SCM on compressive strength of concrete. Although the relationship between compressive strength and sludge substitution differs between reports, the overall trend is a reduction in compressive strength with increasing amount of sludge addition. To ensure sufficient mechanical strength, it is recommended that sludge substitution should be limited to 15% as a conservative estimate, in order to meet relevant standards.

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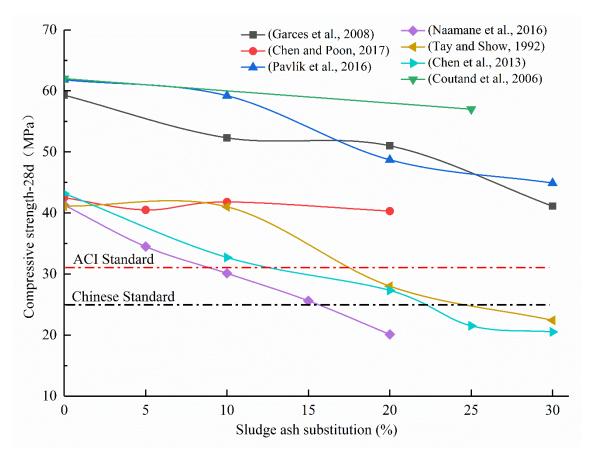


Fig. 4. Effect of incinerated sludge ash used as SCM on compressive strength of concrete. Data from Garces et al. (2008), Chen and Poon (2017), Pavlík et al. (2016), Coutand et al. (2006), Tay and Show (1992), Chen et al. (2013), Naamane et al. (2016). The dashed lines represent the minimum compressive strength requirements for concrete durability according to ACI standard (ACI-318, 2008) and Chinese standard (CCES, 2005).

The pozzolanic activity of sewage sludge ash is highly related to its grain fineness, in which the finer ash particles show higher activity in favour of the strength of mortars (Lin et al., 2008). In addition, mechanical milling also improves the pozzolanic activity of sludge ash as a consequence of a suitable reactive surface available (Donatello et al., 2010). However, the Blaine fineness of sludge ash reached almost 1000 m²/kg after 60 min of grinding, which did not increase significantly for longer grinding times (Dhir et al., 2017b). Generally, sewage sludge ash has a higher specific surface area than sewage sludge after the incineration (Coutand et al., 2006). In addition, the heating or incineration temperature has an effect on the characteristics of sewage sludge ash (Lin et al., 2006; Liu et al., 2018b; Oliva et al., 2019). The results of a study by Lin et al. (2006) indicated that the

porosity of the sewage sludge ash samples decreased when the incineration temperature was increased from 600 °C to 900 °C and a significant decline was observed in the 900-1000 °C in agreement with the variation trends of its water absorption and bulk density.

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Oliva et al. (2019) indicated that the lower incineration temperatures and longer incineration time could increase the specific surface area of incinerated sewage sludge ash related with its cementitious performance. They reported that the blending pastes with 20% substitution of cement by sludge ash showed higher compressive strength than the pastes contained 56% ordinary Portland cement and 44% natural pozzolana, while the strength value was still lower than that of the reference i.e. 100% Portland cement pastes at the same water/cement ratio. In the work of Wang et al. (2017), the properties of cement paste with co-combustion ash consisting of sewage sludge ash and rice husk ash were studied, including hydration characteristics, mechanical properties, freeze-thaw durability, and environmental performance. The results showed that the addition of co-combustion ash as a 30% replacement of the cement inhibited the early hydration process and reduced the compressive strength of sample. However, similar to the study of Cyr et al. (2007), the 7-day and 28-day strength of specimens increased as a result of the potential pozzolanic activity of sludge ash. The study of Naamane et al. (2016) indicated that the pozzolanic activity of sewage sludge reached its maximum at a calcination temperature of 800 °C. The organic matter (especially fatty acids) in sewage sludge negatively affected the compressive strength and prolonged the hydration degree of mortars. A similar conclusion was proposed by Rodríguez et al. (2010). However, the compressive strengths in 90 days became superior to the control mortar, for a replacement ratio of 15%. Pavlík et al. (2016) treated the sludge thermally at a temperature of 700 °C. They suggested the feasible dosage of sewage sludge treated thermally used in cement blend was limited to 10% by weight due to the relatively high content of chlorides and alkalis.

In view of the fineness and lightweight characteristics, sewage sludge ash may be applied in concrete as fine aggregate to replace sand or LWA (Chiou et al., 2006; Lynn et al., 2015). Kosior-Kazberuk (2011) utilized the ash derived from sewage sludge incineration in concrete as a partial

replacement of natural LWA. Their test results showed that the waste aggregate played an important role in mechanical and physical properties of concrete and the acceptable replacement level for structural applications was up to 25% of natural aggregate volume with a compressive and flexural strength of 34 MPa and 6 MPa, respectively. In the work of de Lima et al. (2015), the sand was replaced by sludge ash with a proportion from 0 to 15%. The results showed that there was no obvious variation in compressive strength, porosity and water absorption of mortars with up to 10% of sludge ash, except with a loss of workability for fresh slurry. With the porous and lightweight properties, Wang et al. (2005) used incinerated sludge ash in concrete as LWA to improve the thermal conductivity of concrete. The results demonstrated that the thermal conductivity was decreased with the addition of sludge ash as the porous structure had better heat insulation property, but this improvement was along with the expense of compressive strength. Furthermore, Baeza-Brotons et al. (2014) used sewage sludge ash as a raw material to manufacture concrete blocks. Their results showed that the density value decreased and the water absorption increased when the amount of sludge ash was increased in blocks which was closely linked to the low density and porous structure of the ash particles. Overall, the block sample with 10% replacement of sand by sludge ash showed the best performance in terms of density, absorption, capillarity and mechanical property such as compressive strength compared to the control sample.

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By reviewing relevant literature, it is apparent that the use of sewage sludge ash alone as SCM in mortar or concrete results in a low mechanical strength due to the very slow hydration reaction. Hence, the utilization of sewage sludge ash as cementitious material for the fabrication of concrete requires other activators to improve its performance (Abdalqader et al., 2016). For example, the addition of several pozzolanic minerals such as blast furnace slag (BFS) (Rashad et al., 2016), metakaolin (Cyr et al., 2012), fly ash and lime (Lu et al., 2008)) and alkali activator (Suksiripattanapong et al., 2015) can enhance the performances of sludge ash-based concrete. Chakraborty et al. (2017) studied the performance of sludge ash-based mortar incorporating waste pozzolanic minerals (quicklime and BFS) and alkali activator. At first, the sewage sludge was

incinerated on fluidized bed at 850 °C to produce sludge ash, and the quicklime and BFS were used aiming to increase CaO/SiO₂ ratio and pozzolanic reactivity of mixtures. Based on their results, the mortar fabricated using 70% alkali activated sludge ash, 20% quicklime and 10% BFS showed the best mechanical properties with 31.3 MPa compressive strength, 3.9 MPa flexural strength and 1.61 GPa flexural modulus. In a study by Chen et al. (2018a), ground granulated blast-furnace slag (GGBS) was used to fabricate geopolymer pastes in which sludge ash was used as a precursor. The results revealed that some crystalline minerals such as quartz and hematite in the sludge ash took part in the geopolymerization process and the main products were a combination of both C-A-S-H (calcium aluminate silicate hydrate) and N-A-S-H (sodium aluminate silicate hydrate) gels. The maximum compressive strength of the geopolymer pastes containing 50% sludge ash and 50% GGBS was up to 32.8 MPa at 28 day. In other studies, the best result was achieved at a sludge ash/GGBS ratio of 1:1 (Bai et al., 2003) or 7:1 mixed with another 2 parts quick lime (Chakraborty et al., 2017) as a result of the different characteristics of raw materials. Furthermore, Istuque et al. (2019) utilized the sludge ash as a precursor in the production of metakaolin-based geopolymer. The highest compressive strength of geopolymer mortar reached 50.8 MPa at 180 days cured at 25 °C while a strength retrogression occurred when cured at 65 °C after 7 days. The difference of hydrated products led to this phenomenon in various curing temperature for which the main product of the geopolymer mixture was N-A-S-H type gel at ambient temperature whilst a crystalline phase of zeolite formed at 65 °C (Istuque et al., 2016).

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Based on the studies presented, it appears that sewage sludge ash is more suitable than raw sludge to be used as SCM. The organic matter pyrolysis and the formation of activated minerals in incineration process contribute to the pozzolanic activity of sewage sludge ash. However, excessive amount of ash in cement-based material still affect mechanical properties of products. The amount of sludge ash addition should be limited strictly to 15% to guarantee the quality of sludge-derived product. Furthermore, the addition of other pozzolanic minerals such as blast furnace slag, metakaolin, fly ash and lime improve the performances of sludge product. Through this practice, an eco-friendly

alternative way was provided for comprehensive utilization of all kinds of solid wastes. Further research is needed to generate insights into the potential synergistic-complementary effects among different solid waste materials including sewage sludge, so that high value-added utilization through optimized combination on the basis of their chemical and phase compositions can be implemented.

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3.3. Bricks, roof tiles and ceramic materials

Due to the similarity in the oxide components, sewage sludge and incinerated sludge ash are extensively used as a partial replacement for clay in the fabrication of bricks, roof tiles and ceramic materials. Tay and Yip (1987; 1989) combined the sludge with clay in the production of bricks for construction utilization. In comparing the properties of two types of bricks produced individually by dried sludge and sludge ash fired at 600 °C, they found that the compressive strength of bricks with sludge ash was higher than that of samples with dried sludge at the same dosage. The high content of organic matter in raw sludge resulted in this reduction of strength and a high shrinkage of bricks in firing process. Furthermore, the compressive strength of brick samples with 10% dried sludge and 10% incinerated sludge could reach up to 70% and 98% of the value for the normal clay bricks, respectively. A similar result was demonstrated by Liew et al. (2004) who showed that the compressive strength of sludge-amended clay bricks with the addition of 10% sludge was decreased by 44% as compared with the control specimens, although they still acquired the minimum limits set by the Malaysian Standard. Ukwatta et al. (2015) indicated that organic matter in sludge was a dominant factor affecting sludge use in bricks production. In order to eliminate the impact of organic substance, Lin and Weng (2001) incinerated sewage sludge in a combustion chamber at 800 °C at first and then used incinerated ash to manufacture bricks. According to their results, the increasing amount of ash in the brick decreased plastic index and dry shrinkage and increased the water absorption. However, the compressive strength of bricks firstly increased and then decreased with the increasing content of sludge ash, and the optimal amount of ash added was about 20% of clay by weight. In another study, Weng et al. (2003) indicated that the brick quality depended on the sludge proportion and firing temperature since these two factors affected the shrinkage, water absorption, and compressive strength of bricks significantly. The results presented that the bricks fired at 880-960 °C with an addition of 10% sludge performed as the same as normal clay bricks. The bricks strength met the requirements of the Chinese National Standards when the addition of sludge was up to 20%. In conclusion, sewage sludge has a significant effect on the workability and mechanical property of bricks, therefore the amount of sludge added should be limited to ensure good quality of bricks, for example, at 5% (Kadir et al., 2017) or 9% (Wang et al., 2012).

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In the study by Chiang et al. (2009), rice husk was added for the co-combustion with sludge to fabricate lightweight bricks. With an increased amount of rice husks added up to 20%, the open porosity of bricks increased from 5% to 38% which led to an increase in water absorption and decrease in bulk density and thermal conductivity property. Generally, low thermal conductivity could save the building's energy use. Similarly, fly ash and oven slag were also used incorporating with sludge for brick production (Esmeray and Atis, 2019). Concluded from the microstructure observation, fly ash and slag were beneficial to the dense framework of brick whereas waste sludge had a negative effect. Furthermore, Zhang et al. (2016) produced new environment-friendly bricks composed of 85% lake sediment, 10% cinder and 5% sewage sludge. By observation from SEM micrographs of manufactured brick samples, the incineration of organic matter in sludge resulted in more µm-scale pores and large macro defects on the surface of brick. However, the properties of all brick samples such as compressive strength, water absorption and freeze-thawing resistance still met the Chinese standard requirement. In the work of Doh et al. (2018), eggshell was ground into powder and mixed as partial cement replacement with sewage sludge ash, cement and sand to fabricate mortar bricks. Interestingly, the brick sample containing 10% sludge ash and 5% eggshell showed the best mechanical performance with a compressive and flexure strength of 38.02 MPa and 7.5 MPa which was 1.72 times and 1.14 times of the control bricks, respectively.

Chen and Lin (2009) studied the effects of nano-SiO₂ on the property of tile specimens produced by sewage sludge ash replacing clay from 0% to 50%. They found that nano-SiO₂ additive

reduced water absorption and increased the bending strength of tiles as a result of an improvement on the crystal structures of tile bodies. This positive effect was also confirmed in a study by Lin et al. (2007). In addition, the kiln temperature was suggested to be an important factor dominating the tiles quality as higher sintering temperature contributed to a denser structure in tile body. In another study, Lin et al. (2008) used sewage sludge ash and glaze with different colorants to manufacture tile specimens. Their results showed that the properties of tiles, for example the bending strength, abrasion and acid-alkali resistance, were improved by glaze and the samples with red colorant performed the best properties. In the work of Amin et al. (2018), a pre-pressure at 30 MPa was loaded on tile samples in the fabrication process with dried municipal sewage sludge. The maximum amount of sludge was limited to 7% according to the water absorption requirement (< 10%) of ISO standards (Amin et al., 2018). Furthermore, Cusidó and Soriano (2011) utilized the transformed sewage sludge after ceramization process to manufacture a new pelletized ceramic material which could be used to replace expanded clays in the building or agriculture industry. Compared with the circumstance of incineration, the ceramization process demanded higher temperature (1050 °C) and longer time (12 h). Lightweight material had a microstructure with open porosity and low thermal conductivity, and the leaching tests revealed no adverse effect on environment and human health. Recently, Qi et al. (2010) reported an invention of ultra-lightweight ceramics with a bulk density of 330.80 kg m⁻³ manufactured by dehydrated sewage sludge and clay. The mixing raw materials were preheated at 400 °C for 20 min firstly and then sintered at 1150 °C for 10 min. The aim of preheating process was to remove the absorbed water and structural water and lead to the carbonization of organic matter in sludge. A smooth and porous surface and framework than other ceramics was formed in sintering and bloating process contributing to the ultra-lightweight characteristic of this material. Moreover, Zou et al. (2009) investigated the effect of the major oxides (Fe₂O₃, CaO and MgO) in sewage sludge on the characteristics of ceramsites manufactured from residual sludge. The results showed that the optimal contents of Fe₂O₃, CaO and MgO to produce ceramsite were 5-8%, 2.75-7% and 1.6-4%, respectively. More specifically, residual sludges with 6-8% of Fe₂O₃ could produce ceramsite with

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higher strength due to more complex crystalline phases and fewer pores, and lower strength ceramsite with more pores and amorphous phases could be obtained at 5-7% of CaO. However, MgO had a slight influence on the ceramsite characteristics because Mg²⁺ cannot destroy the unity of crystalline structures. In another study by Xu et al. (2009), the effect of (Fe₂O₃+CaO+MgO)/(SiO₂+Al₂O₃) (F/SA) mass ratio on the characteristics of ceramsite was also studied. The results indicated that the optimal F/SA ratio for making ceramsite ranged at 0.175-0.45, in which ceramsite with a F/SA ratio of 0.175-0.275 had desired physicochemical properties such as higher compressive strength and lower porosity, while at the F/SA ratio of 0.275-0.45 the porous surfaces, expanded structures, and complex crystalline phases could be observed in cermsite resulting in the decrease in compressive strength. Therefore, F/SA ratio should be an important parameter to control during the production process of ceramsite.

Similar to the application of sewage sludge in cement clinker production, the high content of organic matter in sludge is not conducive to the strength of sintered brick. Interestingly, nanomaterials and other waste such as nano-SiO₂, rice husk ash, fly ash and oven slag enhance the quality of sintered bricks by improving the crystal structures and microstructure. Furthermore, the characteristics and properties of manufactured ceramic materials can be regulated by optimizing the oxide composition of raw materials. However, the odor produced from decomposition of organic matter in sludge during the sintering process is highly undesirable, which should be controlled in future research.

3.4. Lightweight aggregates

The utilization of sewage sludge as raw material after pelletizing and firing to manufacture lightweight aggregates (LWAs) is an effective alternative approach for sludge disposal in a sustainable way, which can be widely used in building materials or pavement foundation materials. In a study by Suchorab et al. (2016), 10% sewage sludge and 90% clay by weight were mixed and sintered at 1150 °C for 30 min for the production of LWA. The LWA obtained with sludge addition showed a higher porosity, lower density and reduced compressive strength compared with traditional

LWA. In order to modify the high moisture absorptivity characteristic of LWA, a hydrophobic agent (polysiloxanes) was used for impregnation of aggregates in the preparation of concrete. The results indicated that LWA had good adhesion with cement mortar and no apparent defects such as cracks or scratches were observed. The modified concrete presented a low water absorption between 3% and 7% as a result of the hydrophobic impregnation of aggregates. In the research of Franus et al. (2016), LWA containing 10% sewage sludge showed an increase in porosity when sintering temperature elevated from 1100 °C-1150 °C. This phenomenon was caused by the generation of waste gases liberating from decomposition of organic matter in sewage sludge. A similar finding was observed by Tuan et al. (2013), in which aggregates manufactured with 70% sludge and 30% waste glass powder showed more pores when firing temperature increased. In addition, the more content of waste glass in mixed aggregates, the higher the compressive strength obtained.

More solid wastes are incorporated with sewage sludge in the production of LWAs. Lau et al. (2017; 2018) utilized lime-treated sewage sludge, palm oil fuel ash and sodium silicate to fabricate a new LWA. The main chemical compositions of palm oil fuel ash were SiO₂ (59.13%) and CaO (10.9%) which melted and formed a liquid phase filling the voids between the particles in sintering process. This filling effect resulted in denser structure in aggregate matrix and improvement of mechanical property. In the mixture, sodium silicate (Na₂SiO₃) was used as a binder and lowered the sintering temperature as well (Xu et al., 2008a). Sintered aggregates with binder presented a denser aggregate matrix and high crushing strength in contrast with samples without sodium silicate. Moreover, aggregates produced by sludge and palm oil fuel ash with the ratio of 1:1 and binder with 15% by weight of two raw materials showed a similar strength to commercial aggregates (Lau et al., 2017). The workability and mechanical performance of concrete containing the novel type LWA was comparable with normal concrete (Lau et al., 2018). Coal ash (49.55% SiO₂ and 37.4% Al₂O₃) collected from a power plant was also used in a study by Wang et al. (2009). The addition of coal ash reduced pore size of the sintered aggregates and improved compressive strength while the high concentration SiO₂ and Al₂O₃ demanded higher sintering temperature. When the proportion of coal

ash to dried sludge was 18%-25%, LWA preheated at 420 °C for 20 min and then sintered at 1100 °C for 30 min produced a good quality product with low water absorption (< 8%) and high compressive strength (> 19 MPa). Furthermore, washing aggregate sludge and river sediment containing SiO₂, Al₂O₃, Fe₂O₃ and CaO was also utilized to produce LWAs (González-Corrochano et al., 2016; Xu et al., 2013).

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As sewage sludge ash has similar characteristics and compositions to expansive clay, Chiou et al. (2006) used sludge ash as the principal material together with dewatered sewage sludge to produce LWAs. Sewage sludge ash was obtained from the combustion of dried sludge at 900 °C for 3 h and then ground into finer particles. The results showed increasing the amount of sludge enhanced the bloating effect but decreased the shaping rate due to the specific gravity difference of raw sludge and sludge ash. In order to obtain good spherical particles of LWAs, the amount of added sludge was limited to less than 20%. Cheeseman and Virdi (2005) investigated the effect of sintering temperature on LWA characteristics. When sintering temperature was increased from 1020 °C to 1060 °C, the pores in sintered specimens became discontinuous and isolated, and a dense structure was formed resulting in a decrease in water absorption and an increase in compressive strength. With temperature continually raised up to 1080 °C, the closed pores were gathered together to form larger irregular pores leading to a strength retrogression. This significant effect was also discussed by Liu et al. (2018b). As the sintering temperatures was raised from 900 °C to 1125 °C, the bulk density and compressive strength increased and water absorption rapidly decreased from 65% to 5% due to crystalline phases transformation and the improvement of microstructure. In another study, Liu et al. (2018a) studied the effect of SiO₂ and Al₂O₃ on the properties of LWA mainly composed of sewage sludge and river sediment. In the study, sewage sludge and river sediment were mixed at a mass ratio of 1:1, and different doses of oxides (SiO₂, Al₂O₃) were added into the mixture to adjust the ratio of SiO₂/Al₂O₃. Their results showed that LWA with a SiO₂ content of between 30% and 45% and an Al₂O₃ content of between 11% and 19% presented the highest compressive strength and lowest porosity.

It can be concluded that pyrolysis of sludge promotes the bloating effect and pores formation which is conducive to the lightweight characteristic of aggregate. The high calorific value of sludge lowers the sintering temperature hence saving energy consumption and overall cost. The manufactured LWAs containing 20% sludge present good spherical shape and comparable properties to normal LWAs. In addition, the oxide composition of raw materials and sintering condition such as temperature show a significant effect on the characteristic of LWAs due to the variable characteristics of sludge. However, the long-term performance of LWA produced by sewage sludge still needs further research.

4. Durability

The topic of durability will be high on the agenda when sewage sludge is applied to engineering practice as a construction material. Currently, most studies examined the short-term performance of building materials produced by sewage sludge or incinerated sludge ash, such as workability or mechanical strength at early age, while few investigations examined their long-term performance and durability. Therefore, this review focused on the primary factors determining durability performance of fabricated products and clarified the relationship between durability and characteristics of sewage sludge.

It is well known that porosity and permeability are important parameters affecting the durability of concrete. The open pores in concrete provide possible paths for carbonation, water and chlorine corrosion when exposed to humid environment. Compared with traditional mortar or concrete, a significant increase of porosity in blended mixtures with sludge has been reported (Lin et al., 2005; Lynn et al., 2015; Rodríguez et al., 2010; Yen et al., 2011). Yagüe et al. (2005) evaluated the durability of concrete with the addition of 10% dried sludge, through wet–dry cycles, accelerated ageing and accelerated carbonation tests. Test results showed that the compressive strength of specimens decreased significantly with the increasing amount of sludge after being cured for seven months. However, a slight increase of strength was detected when specimens were submerged in 5%

K₂SO₄ solution, due to the formation of ettringite and the crystallization of salts in pores. As a result of high porosity, the samples containing sludge presented larger carbonation depth. When investigating the frost-resistant performance of cement paste containing co-combustion ash of sewage sludge and rice husk, Wang et al. (2017) reported the deterioration of blended paste was demonstrated by serious compressive strength loss after freezing-thawing tests. Interestingly, amorphous minerals content (approximately 40%) in sewage sludge ash after incineration possess high pozzolanic activity and contribute to a long-term development for mortars strength (Cyr et al., 2007).

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In bricks, ceramic materials and LWAs, water absorption and porosity are significant factors determining the durability performance of these sintered products. Fabricated products with low water permeability usually present good durability and resistance to the natural surroundings (Kadir et al., 2017; Liew et al., 2004). However, in most studies related with utilization of sewage sludge, an increase of water absorption and porosity occurred with increasing amount of sludge in building materials production (Chiou et al., 2006; Cusido and Soriano, 2011; Esmeray and Atıs, 2019; Franus et al., 2016). Fig. 5 shows the effect of sewage sludge substitution on water absorption and compressive strength of bricks. A study by Amin et al. (2018) indicated that the water absorption, apparent porosity and mechanical property were functions of the amount of sludge added. The decomposition of organic matter in sludge caused the formation of more open pores in the mixture and resulted in an increase of water absorption. Similarly, a study of Zhang et al. (2016) attributed the strength retrogression of brick to large weight loss of sludge during sintering. Therefore, the amount of sludge added into bricks should be strictly controlled. Furthermore, a frost-resistance test presented that the weight loss of bricks containing 5% sludge was much lower than 2% after 100 cycles and still met the requirement of Chinese specifications. In the opinion of Chiou et al. (2006), sintering temperature had a significant effect on characteristics of aggregates made by sewage sludge ash. The existing pores on the surface of aggregate were gradually encapsulated by glassy phases at high sintering temperature resulting in low water absorption. Meanwhile, crushing strength of sintered aggregates increased steadily with sintering temperature (Lau et al., 2017). In studying the

incorporation of sludge-based LWA in concrete, Tuan et al. (2013) suggested that concrete containing LWA produced by sewage sludge and waste glass had good corrosion endurance according to the surface resistivity test.

The durability of building material is highly related to its microstructure, especially for open or connected pores in primary structure. In cement-based materials, network cross-linking structure of hydration products is destroyed by inert sludge particles due to the dilution effect. Moreover, the decomposition of organic matter in the alkaline environment of cement slurry causes an increase in pores. Therefore, the pretreatment of sewage sludge is essential before blending with cementitious materials. While in sintering process such as the production of brick and ceramics, pyrolysis and volatilization also promote the formation of more open pores in the mixture. The quality of sludge-amended products can therefore meet the relevant quality standards under limited sludge addition.

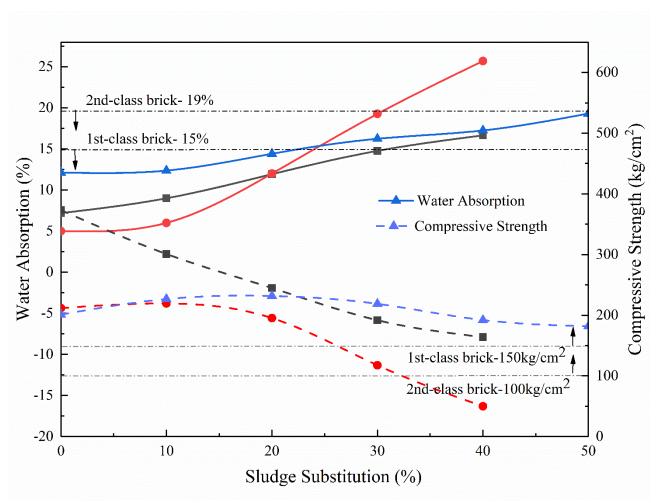


Fig. 5. Effect of sewage sludge substitution on water absorption and compressive strength of bricks, with data in black, red and blue from Juel et al. (2017), Weng et al. (2003), Lin and Weng (2001), respectively. The dot-and-dash lines represent the maximum water absorption and minimum compressive strength

5. Environmental assessment

Due to the nature of sewage sludge, there is a significant concern on the environmental impact of sewage sludge utilization in construction materials. Of particular concern are heavy metal contaminants in sludge which might be introduced to the finished products and cause the problem of secondary environmental pollution. One approach of environmental impact assessment is to conduct leaching tests for raw sewage sludge, sludge ash and sludge-amended products. So far, almost all of the recent experiments show that the leaching concentrations of heavy metals are far below the regulatory thresholds, as summarized in Table 2. However, there is very limited research work on the solidification and stabilization mechanism of heavy metal contaminants in cementitious materials, which is discussed in this part and provides tangible and convincing evidence. These findings should address public concerns and support future applications of this sustainable construction material.

Currently, cementitious material solidification and thermal treatment are two most effective methods for immobilization of heavy metal contaminants (Guo et al., 2017). In cementitious materials, heavy metal contaminants could be embedded and incorporated into the structure of hydration products such as C-S-H with layered structure and needle-like ettringite (Chen et al., 2009; Vespa et al., 2014). On the other hand, heavy metals in sludge might react with components of hydrated products to form precipitates during the cement hydration (Lasheras-Zubiate et al., 2012). Some interactions such as adsorption, precipitation, complexation and encapsulation occurred simultaneously contributing to the immobilization of contaminants (Li et al., 2001). It was found that heavy metals could bind themselves strongly with formed ettringite permanently in a needle-like structure (Dermatas, 1995). Furthermore, the dense structure of hardened cement paste makes a low permeable barrier for preventing the leaching behavior of heavy metals. As a result of the immobilization process for heavy metals, there was no threat posed to the environment when sewage sludge was applied in cementitious material. However, the immobilization process interfered the cement hydration process which retarded the setting time and caused a deterioration in early

compressive strength (Hamood et al., 2017; Lin et al., 2005; Wang et al., 2017).

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Generally, sintering and vitrification are the main methods for thermal treatments of solid waste containing heavy metals and phase transformation of heavy metals occurs in these processes. Sewage sludge usually comprises high contents of SiO₂, CaO, Al₂O₃ and Fe₂O₃, thus a large amount of amorphous and crystal phases is produced during the sintering and vitrification process (Tang and Shih, 2015). The vitreous and crystalline products provide void for heavy metals to be chemically incorporated into the amorphous network and crystal phases or transformed into a new crystal phase (Colombo et al., 2003). For the leachability and toxicity of sludge ash, the pretreatment of the incineration process should convert sludge to a nonhazardous waste (Lin and Weng, 2001). Cyr et al. (2007) detected concentrations of leached contaminants such as zinc, chromium and copper in monolithic and crushed mortars containing 50% sewage sludge ash. Test results presented that crushed mortars released more heavy metals than monolithic mortars due to the more contacting surface area, but both leaching levels were not significantly different compared with reference mortar without residue. When studying the toxic leachability of bricks produced by sewage sludge, Abdul et al. (2004) suggested that the heavy metals contained could be locked inside fired bricks during sintering process and the leaching losses of metals were far below the USEPA regulatory limits. Weng et al. (2003) attributed low contamination leaching level of sludge-amended bricks to the high firing temperature environment which solidified hazardous substance in silicate frameworks. Cusido and Cremades (2012) argued that there was no environmental restriction or any health risk in the production of ceramic products incorporating sewage sludge according to the Netherlands Tank Leaching Test, and the application of sludge as building material should be widely used without restrictions or regulations. Furthermore, a study showed that the leaching contents of heavy metals such as Cd, Cr, Cu and Pb decreased as the sintering temperature was increased from 950 °C to 1050 °C because heavy metals were contained in new crystalline phases at high sintering temperatures (Liu et al., 2018b).

As for the test method for evaluating leachability and toxicity characteristics of sludge-

manufactured products, toxic characteristic leaching procedure (TCLP) is widely adopted whether wastes are in liquid or solid. However, TCLP is carried out under the worst-case test condition so that it would usually overestimate the leaching level for wastes and limit the application of wastes (Halim et al., 2003). Therefore, Liu et al. (2018a) developed a revised method combining TCLP with solid waste-extraction procedure for leaching toxicity (China, HJ 557-2010). Using the revised procedure, they found that the content of SiO₂ and Al₂O₃ in the mixtures had a significant effect on the solidification of heavy metals during the production of LWA. LWA containing 30-45% SiO₂ and 11-19% Al₂O₃ showed the best solidification of heavy metals.

By reviewing leaching test results from the literature, it becomes apparent that there is no immediate environmental threat or human health risk in the production of construction and building materials incorporating sewage sludge. However, the possibility of heavy metals leaching is likely to increase when a large amount of sewage sludge is incorporated to fabricate large-scale construction materials production. In addition, these leaching tests are usually carried out over a short period, leaching behavior under long-term service condition is studied rarely. Therefore, the longer-term safety of sludge-amended construction products should be a priority in future research.

Table 2. Summary of leachability tests for sludge-amended products

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Application	Sludge source	Proportion (%)	Concentration of heavy metals (mg/L)								_ Reference
			Ag	Cd	Cr	Cu	Pb	Zn	Ba	Ni	
Eco-cement	Surface finishing and electroplating sludge	15	ND	ND	0.06	ND	ND	ND	ND	ND	Shih et al., 2005
Eco-cement	STP	12	ND	ND	0.83	ND	0.01	ND	2.02	ND	Lin et al., 2012
Eco-cement	STP	8	ND	ND	ND	ND	0.11	ND	0.06	ND	Lam et al., 2010
Concrete	Textile effluent treatment plant	30	ND	ND	0.73	ND	ND	ND	ND	ND	Rahman et al., 2017
Concrete	STP	20	ND	ND	0.08	ND	0.07	ND	0.30	0.02	Chen et al., 2018b
Bricks	STP	20	ND	ND	0.01	ND	0.32	ND	0.08	ND	Liew et al., 2004
Bricks	Industrial wastewater treatment plant	30	ND	0.01	ND	0.01	0.01	0.12	ND	ND	Weng et al., 2003
LWA	STP	50	ND	0.08	0.13	0.12	ND	0.07	ND	ND	Liu et al., 2018a
LWA	STP	20	ND	ND	ND	0.54	ND	0.40	0.10	ND	Chiou et al., 2006
	TCLP ^a Regulatory Limit		5	1	5	15	5	25	100	25	
	GB 5085.3–2007 ^b Limit		5	1	15	100	5	100	100	5	

⁷⁴⁶ ND: not detected; ^aTCLP: Toxicity Characteristic Leaching Procedure; ^bChinese Standard: Identification Standards for Hazardous Wastes – Identification for extraction toxicity

6. Conclusions

By critically and extensively reviewing published research on the application of sewage sludge and incinerated sewage sludge ash in construction materials, it is concluded that this solid waste can be utilized as: (i) raw material components for the production of eco-cement, bricks, ceramic materials and LWAs through sintering process; (ii) supplementary admixtures in cementitious materials such as pozzolanic component, fine aggregate or filling material. Key conclusions are presented as follows:

- (1) SiO₂, Al₂O₃ and CaO are the main oxides in sewage sludge and sludge ash, which are essential oxide components for cementitious materials. Compared to raw sewage sludge, sludge ash has a higher content of oxides due to the loss of volatile components and decomposition of organic matter after incineration. In addition, high specific surface area and amorphous phase account for high pozzolanic activity of sludge ash.
- (2) As a substituting raw material, sewage sludge could be used safely up to 15% in producing eco-cement which possesses similar mineral components and comparable performance to traditional Portland cement. Excessive amount of sludge substitution causes a deterioration in compressive strength, and an increase in water demand and final setting time of eco-cement paste. Furthermore, the high calorific value of sewage sludge contributes to a fuel substitution for energy conservation.
- (3) The addition of raw sewage sludge decreases the mechanical strength and hinders hydration process of cement or mortar significantly as a result of high organic matter content. CLSM offers an innovative approach to recycle large quantities of sewage sludge where low strength of the structure is acceptable. Sewage sludge ash is widely used as supplementary admixtures with high pozzolanic activity in cementitious materials. With 10-20% of sludge ash in blended mixtures, there is a slight

but acceptable decrease in workability and mechanical strength compared with samples without ash residues. Furthermore, other pozzolanic minerals such as blast furnace slag, metakaolin, fly ash and lime improve the structural performance of sludge-manufactured products.

- (4) As raw feed in the fabrication of bricks, ceramic materials and LWAs, increasing amount of sewage sludge increases water absorption and porosity of sludge-amended products, which are the main factors adversely affecting the long-term performance and durability. In order to produce construction materials of a sound mechanical structure, the maximum dosage of sludge addition should be limited to 20%.
- (5) In cementitious materials, heavy metal contaminants in sewage sludge can be incorporated into the structure of hydration products or react with hydrated products to form stable precipitates. Subsequent sintering process ensures the immobilization of heavy metals by chemically incorporating into the amorphous network and crystal phases or transforming into a new crystal phase. The use of sewage sludge in construction materials therefore serves two purposes: recycling of sludge as a raw material and immobilization of heavy metals, therefore converting a solid waste to valuable products while preventing secondary environmental pollution.

7. Recommendations for future research

Although there have been extensive studies on various utilization methods of sewage sludge in building materials, research remains to be conducted in order to solve many problems and advance this relatively new research field.

(i) Thermal treatment of sewage sludge such as incineration promotes utilization of sludge but consumes additional energy. So far, there has been limited research on raw sewage sludge application in high performance concrete due to its high organic matter content and low pozzolanic activity. As

concrete is the most widely used building material, it represents the most effective way to recyce and convert sewage sludge to high value-added products. Therefore, future research should focus on reducing adverse effects of sludge application in concrete. Firstly, more effective pretreatment (e.g. microwave) for removing organic matter in sludge needs to be explored. Referring sewage sludge as an inert material, further study should improve the mechanical property of sludge-amended cementitious materials by optimizing particle size distribution based on particle dense packing theory. Secondary, comprehensive utilization techniques for sewage sludge with pozzolanic materials should be strengthened to improve cementitious properties of sewage sludge. Finally, it is necessary to develop more effective modified treatment of sludge to improve its physical property, chemical activity and cementitious characteristics (such as alkali-activation), aiming to upgrade the quantity and quality of sludge utilization.

- (ii) For the application of dried sewage sludge and sludge ash in cement or mortar, increased water requirement and workability loss can be overcome by superplasticizers. While few researchers investigate the compatibility of superplasticizers or other polymer additives with sludge, further research should be conducted to optimize experimental conditions with cement additives. Furthermore, the potential synergistic-complementary effect among all kinds of solid wastes need further exploration. The high value-added and comprehensive utilization will be implemented by optimized combination through scientific design methods on the basis of their chemical and phase compositions.
- (iii) The porous nature and high specific surface area of sewage sludge ash particles demand further exploitation in latent application areas. For example, sludge ash can be used as water-retaining LWA or internal curing agent in concrete which can absorb excessive water into its pores at early age, and release water gradually from pores during maintenance subsequently. Moreover, adsorbent effect

of ground ash particles deserves more attention.

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- (iv) Sludge-amended products show comparable short-term performance to traditional building materials at low sludge content. More importantly, their long-term performance and durability need further investigations, in determining heavy metals leaching risk, acid/sulfate attack, carbonation, freeze-thawing and steel reinforcement corrosion.
- (v) For the application of sewage sludge in conventional building materials, essential and corresponding technical criteria or guidelines should be formulated by systematic research to guide further engineering applications.
- (vi) Interesting research should be performed by full valorization of sewage sludge. For example, extracting metal elements from sewage sludge and producing activated carbon absorbents from the organic matter can be completed before sludge ash being used as SCMs.

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