Effect of lime addition during sewage sludge treatment on characteristics of resulting SSA when it is used in cementitious materials

D. Vouk, D. Nakic, N. Štirmer and A. Baricevic

ABSTRACT

Final disposal of sewage sludge is important not only in terms of satisfying the regulations, but the aspect of choosing the optimal wastewater treatment technology, including the sludge treatment. In most EU countries, significant amounts of stabilized and dewatered sludge are incinerated, and sewage sludge ash (SSA) is generated as a byproduct. At the same time, lime is one of the commonly used additives in the sewage sludge treatment primarily to stabilize the sludge. In doing so, the question arose how desirable is such addition of lime if the sludge is subsequently incinerated, and the generated ash is further used in the production of cementitious materials. A series of mortars were prepared where 10–20% of the cement fraction was replaced by SSA. Since all three types of analyzed SSA (without lime, with lime added during sludge stabilization and with extra lime added during sludge incineration) yielded nearly same results, it can be concluded that if sludge incineration is accepted solution, lime addition during sludge treatment is unnecessary even from the standpoint of preserving the pozzolanic properties of the resulting SSA. Results of the research carried out on cement mortars point to the great possibilities of using SSA in concrete industry.

Key words | cement mortar, lime, pozzolanic material, sewage sludge ash, sludge stabilization

INTRODUCTION

Adequate wastewater management implies collecting, transport and treatment of wastewater, but also adequate management of waste generated by the treatment processes. Treatment of wastewater and management of sewage sludge are important issues at the global level. The same issues are becoming increasingly important in Croatia as well, with regard to the fact that the commitments towards the EU have resulted in intensive designing and construction of a great number of wastewater treatment plants (WWTP) larger than 10,000 people equivalents (PE). For WWTP with medium capacities (between 5,000 and 200,000 PE) sewage sludge treatment and disposal costs are nearly 50% of total operating costs for wastewater treatment (Nowak et al. 2003). In most countries (even well developed countries), the problem of adequate disposal of sewage sludge has not been adequately solved, nor is it determined by the rules, instructions or guidelines. As construction of WWTPs is becoming more intensive on a worldwide level, sludge disposal will become a burden to many water utilities managing sewerage and wastewater treatment systems. By 2020, the EU is expected to produce about 13 million tons of dry matter (DM) of sewage sludge per annum (Milieu Ltd 2010; Leonard 2011). Within the framework of sustainable development, recycling of sludge or byproducts of its further management almost completely closes the cycle of wastewater treatment in which only negligible amounts of waste products that need to be disposed in the environment are generated. In selection of the final solution of sewage sludge treatment and its final disposal, the important issue is chemical composition, including nutrients. According to Metcalf & Eddy (2014), control of pH, alkalinity and organic acids content are of prime importance in processes of anaerobic digestion. Further, the content of heavy metals, pesticides and hydrocarbons also requires detailed analyses in cases of sludge incineration or disposal. Considering the energetic (thermal) value of sludge is also extremely important from...
the aspect of thermal processing (gasification, pyrolysis, incineration, wet oxidation) (Pytili & Zabaniotou 2008).

Incineration of sewage sludge considerably facilitates further management of the new product (sewage sludge ash (SSA)), first of all as a consequence of significant reduction of mass and volume of the final by product. Also, incineration destroys toxic organic components, odours are reduced to minimum, further management is facilitated, and power generation is also possible (Tantawy et al. 2012). At the EU level about 22% of generated sludge is incinerated (Lynn et al. 2015). Incineration has arisen as an alternative approach to sewage sludge disposal which provides water utilities a great deal of stability and control over sludge management. If the sludge incineration concept is accepted, considerable quantities of SSA, that also require disposal, are still generated. Donatello & Cheese- men (2015) estimate that at present, at the global level, thermal processing of sludge from WWTP generates 1,700,000 tons of SSA per annum (mainly in the USA, the EU and Japan). Construction of new and reconstruction of existing WWTP will result in constant increase of this number. For example, in Croatia in 2018 there will be treatment plants in operation with the total capacity of 4,500,000 PE, generating the total quantity of stabilized and dewatered sludge of about 250,000 tons per annum. If incineration of sewage sludge is chosen, the production of SSA would be 57,000 tons per annum. The possibility of subsequent recycling and use of SSA greatly depends on its composition, mainly chemical. Likewise, the composition of sewage sludge and SSA depends on the wastewater origin, but also on the technological process of wastewater and sludge treatment (for instance, which level of treatment is applied (secondary or tertiary) or whether lime is added to sludge in the final phase of treatment, etc.) (Nakic et al. 2015).

In accordance with principles of sustainable development, at present there is a growing tendency of recycling of various (waste) materials. In this context, the possibility on recycling and use of SSA in the construction industry, which is a great consumer of raw materials, particularly stands out. This is especially significant in the production of cementitious materials, since SSA exhibits certain pozzolanic activity and the main chemical elements present in Portland cement (Ca, Si, Al and Fe) are also present in SSA (Donatello & Cheeseman 2013; Lynn et al. 2015; Vouk et al. 2015). The amorphous content of SSA ranged from 35 to 75% according to Lynn et al. (2015) which means it is somewhat reactive. Using direct methods, Jam-shidi et al. (2011) determined the pozzolanic activity of SSA to be 37.86%, while Fontes et al. (2004) got the value of 70.53%. According to present research, SSA may be used in cementitious materials as active pozzolanic material (Paya et al. 2002; Cyr et al. 2007; Garces et al. 2008; Donatel-llo et al. 2010; Baeza-Brotons et al. 2014; Nakic et al. 2015) or as inert filler (replacing part of sand or fine aggregate). As stated before, possibilities of using and recycling of SSA greatly depend on its chemical composition, which is related to the sludge origin and the wastewater and sludge treatment technology. Regarding environmental influence of SSA when used in cementitious materials, the leaching behaviour of mortars containing SSA is of the same order of magnitude as that of reference mortar without the SSA residue according to Coutand et al. (2006).

Lime is one of the commonly used additives in the sewage sludge treatment at WWTPs, primarily to stabilize the sludge, especially in cases when sludge is landfilled. Increasing pH value by adding a lime (as CaO) above 12 reduces levels of pathogenic bacteria and viruses in the sludge and controls putrefaction and odours (Metcalf & Eddy 2014). Typical amounts of added lime are about 20–30% relative to the amount of DM of sludge (EPA 1981). This is also the current practice in many WWTPs worldwide as well as in Croatia. However, with the EU accession and restrictions of sludge landfilling, development of a new strategy on sewage sludge disposal in Croatia has started. Although this strategy is still in development, the most frequently mentioned is construction of regional incineration plants. The question that arises is how desirable is such addition of lime if the sludge is subsequently going to be incinerated, and the resulting SSA is going to be used in the production of cementitious materials. The main aim of this paper is to give answers on that question as well as to examine the possibility and feasibility of incorporating of SSA in cementitious materials with emphasis on the influence of the lime addition, whether during sludge stabilization or when incinerated. Lime may also be added during incineration process to reduce the slagging of sludge (Anon 1992). This is particularly important at high operating temperatures (above 900 C) which can result in partial fusion of ash particles and the formation of clinker. SSA produced at WWTP that introduce ferrous salts or lime for sludge conditioning and dewatering contains significantly higher quantities of iron and calcium, respectively. The pH of SSA can vary from 6 to 12, but it is generally alkaline (Cox et al. 2008). Results of physical and chemical characteristics of SSA from WWTP Zagreb (Croatia) will be presented in the paper together with the study analysis of the influence of SSA on physical and mechanical properties of cement mortars.
EXPERIMENTAL RESEARCH

Sludge was collected from WWTP Zagreb (Croatia) that operates with second stage of treatment and uses conventional activated sludge treatment technology. Sludge treatment is based on thickening, anaerobic digestion, lime addition and finally dewatering. For the purpose of this research, sludge without added lime during stabilization was also collected.

SSA used in this research was obtained by sludge incineration in laboratory conditions using electrical muffle furnace. Collected sludge was additionally dried at 105 °C to constant mass and a DM content of about 90% and then incinerated at temperatures of 800 °C and 1,000 °C which are chosen on the basis of state-of-the-art (Donatello & Cheeseman 2013; Vouk et al. 2015). Also, sludge that was stabilized with lime was incinerated with 20% of extra lime (by mass) during sludge incineration. This was done in order to analyse the influence of an additional amount of lime on the pozzolanic characteristics of the resulting SSA. The intention was also given to the aspect of reducing the slagging of sludge. All generated SSAs were in a form of granular material and therefore additional grinding was required. The grinding was performed in an electric grinder for about 30 s for each batch.

SSA obtained in this study was used as a replacement for part of the cement (10% and 20% by mass, which is chosen based on the results of previous research) in mortars. Determination of the chemical composition of generated SSA was based on the analysis of the share of individual oxides by the Standard EN ISO11885:2010.

CEM II/B-M (S-V) 42.5N cement and dolomite sand (0/4 mm) were used to prepare mortars. The water–binder ratio was 0.50. Ordinary tap water at room temperature was used. Batches were prepared in a 5 L mixing bowl, with 2,027 g of cement, 507 g of SSA, 1,267 g of water and 7,508 g of sand (water : cement : sand = 0.5 : 1 : 3). In reference mixture, the whole binder is cement, without the addition of SSA (total cement content 2,534 g). Batches were mechanically mixed. Flow table spread (FTS) was given in accordance with EN 1015-3:1999. Air content was determined using ‘pressure method’ on the basis of EN 1015-7:1998. Specimens for testing of flexural and compressive strength were prepared in the form of 4 × 4 × 16 cm prism monoliths. Specimens were mechanically tested after 1, 7 and 28 days of curing. For each mortar and curing age, three specimens were tested. Mortar samples were tested in bending, and the two resulting pieces were tested under compression. The mechanical tests were performed in accordance to the EN 1015-11:1999 standard. Nine specimens were made from each mix using steel moulds, with three tested for flexural strength at each curing age (1, 7 and 28 days). Compressive strength was tested on the resulting halves (i.e. a total of six samples at each curing age).

RESULTS AND DISCUSSION

SSA characteristics

The type of wastewater and the additives used during sludge treatment at the WWTP influence the composition of SSA greatly (Lopes et al. 2005; Chen et al. 2015). The main crystalline phases present are quartz (SiO2), calcium phosphate (Ca3(PO4)2) and haematite (Fe2O3). Also, SSA can primarily be considered amorphous due to high temperature incineration (Perez-Carrion et al. 2014). SSA contains significant levels of phosphate, usually between 10 and 20% by mass, in the form of P2O5 (Donatello & Cheeseman 2013). Chemical composition of obtained SSA in the term of main present oxides is given in Figure 1.

Pozzolanic activity can initially be expected from the SSA obtained since its chemical composition has the largest share of SiO2, CaO and Al2O3. Significant shares of CaO may be attributed to considerable quantities of lime added
in the process of sludge treatment (before dewatering phase) and in line with the expected SSA without lime that has the lowest proportion of this oxide while the SSA obtained by co-incineration of sludge with lime contains the largest shares of CaO (up to 59%).

Densities of all obtained SSAs were somewhat lower than the density of the cement (about 15% lower) and the mean density of all SSA samples was 2.65 g/cm³, with standard deviation of 0.11. Mass reduction during incineration was the most significant for the sludge without added lime, and least expressed for co-combustion of sewage sludge with additional lime. As initially expected, with an increase in combustion temperature, mass reduction of sludge (i.e. resulting SSA) is more expressed. Average mass reduction at combustion temperature of 800 °C was about 46%, and about 50% at 1,000 °C for sewage sludge without lime. For co-combustion (sewage sludge + additional lime) average mass reduction at combustion temperature of 800 °C was about 50%, and about 54% at 1,000 °C. Results for the sewage sludge that contained lime which was added during the sludge treatment (before dewatering phase) were in between results of the above mentioned two types of sewage sludge.

According to literature review, the particle size of SSA ranges from 1 to 100 μm, with mean diameter value of about 26 μm (Coutand et al. 2006; Yusuf et al. 2012). For all types of SSA obtained from the sludge from WWTP Zagreb (Croatia), most of the particles were between 48 and 63 μm (after subsequent grinding). The particle size of analyzed SSA ranges between 5 and 500 μm (Figure 2), with a mean diameter around 50 μm. Less than 2% of particles are less than 10 μm.

According to the results, in terms of particle size, SSA could be classified as silt with a significant proportion of particles with the size of sand.

### Characteristics of fresh mortar

Fresh state characteristics are important for future application of new materials and have major influence on the behaviour of cementitious materials in hardened state.

All mixtures showed similar values with regard to density of fresh mortar (on par with the density of reference mix) and minimum differences among mixtures were noticed (maximum deviation was within ±2% relative to the density of reference mix) (values of density in fresh mortars were obtained with an average value of 2.3 g/cm³). With the addition of the SSA, a slight increase in the temperature of the fresh mortar was observed (2–4 °C increase compared to reference mixtures). This was expressed more for SSA that contained higher CaO content (i.e. SSA obtained by co-combustion of sewage sludge and additional lime). Although, higher reactivity of those mixtures could be primarily because of the so called ‘filler effect’, it could also be partially attributed to somewhat higher CaO content.

Furthermore, all SSA mixtures have higher air content compared to the reference mixture which is less significant for mixtures containing SSA without lime (average increase in air content 20–30%), and much more expressed for mixtures with SSA containing additional amounts of lime added either during sludge stabilization or during sludge incineration (average increase in air content about 85–90% compared to the reference mixture) (Figure 3). Independently, increase of incineration temperatures has positive influence on the amount of air inside of the mix, i.e. air content is reduced. This is in line with determined physical
characteristics of SSA, where it was demonstrated that an increase of incineration temperatures increases the specific density of the SSA. Higher specific density assures better packing of particles inside of the composite.

It is well known that incorporation of large percentage of very fine particles causes higher water demand and without additional corrections of mix design implicates lower consistency of mix (Bjegović & Štirmer 2015). Consequently, an increase of water demand of mortars containing SSA as a cement replacement was noticed. The water demand of SSA is related to high specific surface area of the grains, which are mainly composed of small sintered particles. In accordance with that, mixes containing higher amounts of SSA showed significantly lower FTS values.

The mortar workability results show decreasing trend in FTS values with increasing SSA share in mortars for all mixtures. There is also a trend of higher decrease in the workability of mortars with SSA obtained at a higher incineration temperature (1,000 °C) (Figure 4). Generally, the best workability was obtained for mixtures with SSA that do not contain any lime (Figure 4) and it is on par with the workability of the reference mixture. Mixtures containing SSA with lime showed an average decrease of workability compared to the reference mixture, of about 10–15% for the addition of 10% of SSA and 20–25% for the addition of 20% of SSA. The decline in workability is nearly linear, and the average rate of decrease in workability calculated is 10% for every 10% of SSA. This negative effect may be compensated for with addition of alternative chemical additives such as plasticizers and superplasticizers.

**Microstructure of hardened mortars**

Scanning electron microscope (SEM) analyses of cement mortars containing the SSA with a share of 20% show characteristic grains of granulated materials (Figure 5) that indicate the existence of chemically different phases. This was also confirmed by X-ray diffraction (XRD) and chemical analysis. Results of XRD analysis showed the significant presence of an amorphous phase and of many crystalline minerals, the main ones being quartz and cristobalite (SiO2), muscovite, anhydrite (CaSO4) and calcite (CaCO3). Also, it was found that porosity is higher for samples with higher SSA content.

Many micro-pores, seen in microstructure of hydrates of cement mortars, are indicating that the interface between the SSA and calcium-silicate-hydrate (C-S-H) gel was bonded together well to produce a microstructure with fine and non-uniform particles. Although differences in chemical composition of different samples were observed by SEM, at these magnifications different chemical compounds cannot be specified. Despite the fact that...
Figure 5(a) and 5(c) apparently seem alike, it is important to notice that sample with additional lime (Figure 5(c)) has somewhat larger particles and higher porosity.

All samples were characterized by poorly porous sheet material with a greater or lesser degree of heterogeneity embedded in a matrix of isotropic particles with a greater or lesser degree of agglomerating.

**Mechanical properties**

Although the general trend of slight reduction in compressive and flexural strength was observed with increased SSA addition, some mixtures containing SSA have shown equal or even higher strengths than the control mixture (Figure 6). Although certain differences exist, there were no significant discrepancies in the results for the mechanical properties of mortars prepared with SSA obtained at different incineration temperatures (800 °C and 1,000 °C).

It can be seen (Figure 6) that, although relatively high strengths are obtained, the increase of replacement level of cement with SSA led to a decrease in compressive and flexural strengths for all hydration times. Anyway, the gaps between SSA and reference mortars, which are most pronounced for the 1-day strengths, reduce significantly over time indicating that there is a certain reaction. This is consistent with the conclusions of Paya et al. (2002) that the formation of hydraulic products from the pozzolanic reaction between SSA and cement takes place mainly during medium curing times which means that SSA could be considered as medium-reactive pozzolanic material. Similar conclusion that the short-term negative effect of SSA was strongly reduced after 7 days was given by the Cyr et al. (2007).

Considering all the samples that have been tested, as the best with respect to the mechanical characteristics, mix with 10% of SSA (without lime) obtained at 800 °C, stands out. This mix has reached 95% of the 28-day flexural strength of the reference mix, and even 98% of its compressive strength. Also, for the hydration period of 7 days, its flexural strength even exceeded the one of the reference mixture. On the other hand, the weakest results were obtained for a mixture with 20% of SSA (without lime) obtained at 1,000 °C, 74% of the flexural strength of the reference mixture and 84% of its compressive strength.

The weakest results within the mixtures where SSA without lime was used were obtained in the early stages of hydration (1-day strengths). A smaller share of CaO in this type of SSA is assumed to be one of the main reasons for somewhat slower reaction.

Overall, the average decrease of compressive strength values is less than 8% with the addition of 10% of SSA,
CONCLUSIONS

In line with the increasing attention given by many water utilities regarding the incineration of sewage sludge there has been many researches on recycling and recovery of SSA lately. Many of them focused on the use of SSA in a production of cementitious materials. Recycling of SSA in cementitious materials provides both, environmental and economic benefits, primarily through conservation of raw materials, but also through the reduction of waste being sent to landfills.

Results of investigations carried out on cement mortars show that samples in which part of cement is replaced by SSA have most of the properties similar to those of reference samples. Mortars prepared with limited SSA additions (up to 20%) exhibit similar properties to control mixtures that do not contain SSA. All three types of analyzed SSA (without lime, with lime added during sludge treatment and with extra lime added during sludge incineration) yielded nearly same results. So, it can be concluded that if sludge incineration is accepted, lime addition during sludge treatment is unnecessary even from the standpoint of preserving the pozzolanic properties of the resulting ash when it is subsequently used as a partial cement replacement in the production of cementitious materials. Eventually, when evaluating all possible benefits from usage of SSA, lower energy consumption when sludge is incinerated at lower temperatures should also be considered as an additional prerequisite for achieving economic and environmental sustainability.

In accordance with the principles of energy efficiency and minimum environmental impact, sludge incineration with the use of SSA in the construction industry might be considered as acceptable solution. Unique legislation on this issue (i.e. at EU level) would also contribute to its wider acceptance.

ACKNOWLEDGEMENT

This work has been fully supported by the Croatian Science Foundation under the project ‘927 – Reuse of sewage sludge in concrete industry – from infrastructure to innovative construction products’.

REFERENCES


Bjegović, D. & Stirmr, N. 2015 Theory and Technology of Concrete. University of Zagreb Faculty of Civil Engineering, Zagreb, Croatia.

Chen, M., Blanc, D., Gautier, M., Mehu, J. & Gourdon, R. 2013 Environmental and technical assessments of the potential utilization of sewage sludge ashes (SSAs) as secondary raw materials in construction. Waste Manag. 33, 1268–1275.


Fontes, C. M. A., Barbosa, M. C., Toledo Filho, R. D. & Goncalves, J. P. 2004 Potentiality of sewage sludge ash as mineral...


First received 15 June 2016; accepted in revised form 17 November 2016. Available online 8 December 2016