Evaluation of the pozzolanic activity of uncontrolled-combusted sewage sludge ash

Danilo Bordan Istuque[1], Lucia Reig[2], Lourdes Soriano[3], Maria Victoria Borrachero[4], José Luiz Pinheiro Melges[5], Jorge Luis Akasaki[6], Jorge Juan Payá Bernabeu[7], Mauro Mitsuuchi Tashima[8]

[1] UNESP - Universidade Estadual Paulista "Júlio de Mesquita Filho", Faculdade de Engenharia de Ilha Solteira. MAC – Grupo de Pesquisa em Materiais Alternativos de Construção. Ilha Solteira-SP, Brazil.

[2] EMC, Universitat Jaume I, Av. de Vicent Sos Baynat s/n, 12071 Castello´ de la Plana, Spain.

[3] ICITECH – Instituto de Ciencia y Tecnología del Hormigón. Universitat Politècnica de València (UPV). Valencia, Spain.

[4] ICITECH – Instituto de Ciencia y Tecnología del Hormigón. Universitat Politècnica de València (UPV). Valencia, Spain.

[5] UNESP - Universidade Estadual Paulista "Júlio de Mesquita Filho", Faculdade de Engenharia de Ilha Solteira. MAC – Grupo de Pesquisa em Materiais Alternativos de Construção. Ilha Solteira-SP, Brazil.

[6] UNESP - Universidade Estadual Paulista "Júlio de Mesquita Filho", Faculdade de Engenharia de Ilha Solteira. MAC – Grupo de Pesquisa em Materiais Alternativos de Construção. Ilha Solteira-SP, Brazil.

[7] ICITECH – Instituto de Ciencia y Tecnología del Hormigón. Universitat Politècnica de València (UPV). Valencia, Spain.

[8] UNESP - Universidade Estadual Paulista "Júlio de Mesquita Filho", Faculdade de Engenharia de Ilha Solteira. MAC – Grupo de Pesquisa em Materiais Alternativos de Construção. Ilha Solteira-SP, Brazil. *Corresponding author. Email: mauro.tashima@unesp.br

Abstract

The waste management is a crucial issue which should be solved by the modern society. The generation of sewage sludge is increasing annually due to the urbanization and improvement on the sanation system of cities. In this sense, the building and construction sector emerges as a solution for waste disposal due to the huge volume of materials that can be absorved by this sector. This paper evaluates the pozzolanic activity of sewage sludge ash (USSA) obtained following an uncontrolled-combustion process, a simple and economic procedure. Compressive strength of Portland cement/USSA mortars with different percentage of USSA (5–25 wt.%) were evaluated, as well as calcium hydroxide/USSA (CH/USSA) and Portland cement/USSA (PC/USSA) pastes were chemically and physically characterised through TG/DTG, FTIR, XRD and SEM analyses. The obtained results revealed that the increment on the Portland cement replacement by USSA is associated to an increasing on the compressive strength of mortars. For mortars containing 25% of USSA, the increment of compressive strength yielded values of 27%, 16% and 7% after 7, 28 and 90 curing days, respectively. According to the microstructural analysis, the increment on the compressive strength can be attributed to formation of hydrated products (C-S-H, C-A-S-H, C-A-H) by the pozzolanic reaction of USSA.

Keywords: Sewage sludge ash (USSA); Pozzolan; Fixed portlandite; Waste valorisation; Uncontrolled combustion.

Introduction

Increasing amounts of sewage sludge, a waste generated in wastewater treatment plants, are yearly generated, which is mainly attributed to the urbanisation and improved sanitation systems of the cities. According to Krüger and Adam (Krüger and Adam 2015), 30 million tons/year of sewage sludge are generated by Europe, North America, and Japan (the sum of all of them). Kelessidis and Stasinakis (Kelessidis and Stasinakis 2012) also pointed out that it is expected that by 2020 the production of dry sewage sludge in the European Union will exceed 13 million tons. The large volume of sewage sludge generated prompted the development of technological plants to incinerate this waste while generating energy (Abuşoğlu et al. 2017; Donatello and Cheeseman 2013). Although this significantly reduces the volume of waste, the ash resulting from the process must also be adequately managed (Kliopova and Makarskienė 2015; Liu et al. 2014; Wang et al. 2012). According to Donatello and Cheeseman (Donatello and Cheeseman 2013), the estimated global production of sewage sludge ash (SSA) is 1.7 million tons/year, being mainly produced in the USA, the European Union, and Japan. The global production of SSA is expected to increase in the future since countries such as Belgium, Portugal, Ireland, or Spain support the incineration of sewage sludge (Kelessidis and Stasinakis 2012). Thus, the reuse and valorisation of SSA is of great interest and contributes to a circular economy since, in agreement with Smol et al. (Smol et al. 2015), it diminishes the amount of waste generated, while adding new value to it.

In this sense, previous studies successfully recovered phosphorous from SSA (Krüger and Adam 2015), while other research used SSA as a raw material to produce different construction products, such as blended Portland cement, pastes, mortars, bricks, tiles, ceramics or glass (Baeza-Brotons et al. 2014; Chen and Poon 2017; Dyer et al. 2011; Lin et al. 2007; Monzó et al. 2003; Perez Carrion et al. 2013; Smol et al. 2015; Tarrago et al. 2017; Tashima et al. 2017; Yusuf et al. 2012; Zhou et al. 2019). Reusing SSA in these applications was possible due to its pozzolanic behaviour, which is affected by the amorphous content of

60 SiO_2 and Al_2O_3 (Cyr et al. 2007; Garcés et al. 2008; Lynn et al. 2015) apart from the temperature and time
61 of the sewage sludge incineration (Naamane et al. 2016; Oliva et al. 2019).

62 All the studies performed until now had in common that the temperature and time used to incinerate the
63 sewage sludge were controlled (Chen et al. 2013; Cyr et al. 2007; Garcés et al. 2008; He et al. 2017; Li et
64 al. 2019, 2017; Monzó et al. 2003; Naamane et al. 2016; Yusuf et al. 2012). However, controlled
65 incineration processes usually require large and technological plants, which are initially expensive (Xin-
66 gang et al. 2016). As explained by Kelessidis and Stasinakis (Kelessidis and Stasinakis 2012), when the
67 sewage sludge cannot be incinerated, the most common alternative is disposing of it in landfills. Therefore,
68 using simple and economic methods of incineration would allow reusing this waste anywhere, regardless
69 of the existence of incineration plants. The research reported here aimed to develop a simple route to
70 incinerate sewage sludge, and to evaluate the reactivity of the resulting ash, which was called uncontrolled-
71 combusted sewage sludge ash (USSA). The USSA was characterised (XRF, XRD, PSD, FTIR, and SEM
72 analyses), and its pozzolanic behaviour was assessed using calcium hydroxide/USSA pastes (CH/USSA;
73 TG/DTG analyses), Portland cement/USSA pastes (PC/USSA; TG/DTG, XRD, FTIR and SEM analyses),
74 and PC/USSA mortars (compressive strength development).

75

76 ## Materials and Methods

77 ### Materials

78 Dewatered sewage sludge with relative humidity of 77%, approximately, was collected in the São José do
79 Rio Preto wastewater treatment plant (São Paulo, Brazil). Brazil. High purity calcium hydroxide (> 95% of
80 $Ca(OH)_2$ – "CH") was used to prepare CH/USSA pastes. The Brazilian Portland Cement CP V ARI (PC)
81 used to prepare pastes and mortars presented a clinker content greater than 95% and did not contain
82 pozzolanic additions. Siliceous sand from Castilho city (São Paulo – Brazil), with a particle diameter lower
83 than 2.36 mm, a fineness modulus of 2.12, and a specific gravity of 2.64 g/cm³ was used to prepare the
84 PC/USSA mortars.

85

Methods

Incineration of sewage sludge

The process followed to produce USSA is shown in Fig. 1. Firstly, around 3 cm layers of dewatered sewage sludge were dried by exposing them to solar radiation for four days. Secondly, the dried-granular sewage sludge was incinerated in an uncontrolled-combustion cylindrical chamber (200-litre volume). About 20 kg of dried-granular sewage sludge were put in the chamber, and free air circulation was initiated. To allow the combustion to initiate, gas was supplied in the bottom of this chamber during the first minute. The complete combustion of the dried-granular sewage sludge occurred by the propagation of the heat from the bottom to the top. This process was repeated several times until the amount of ash required to perform the study was produced. The temperature of the uncontrolled-combustion of the sewage sludge was monitored with a thermocouple installed inside the oven. As shown in Fig. 2, after 3 hours of combustion, a maximum average temperature of approximately 774°C was reached. The incineration of sewage sludge with a temperature above 500°C is important to loss of volatile components and decomposition of the organic matter, that in high content affects the mechanical properties of cementing materials (Chang et al. 2020). Furthermore, according to Naamane et al. (Naamane et al. 2016), as nearer to 800°C occurs the calcination of the sewage sludge, higher is the pozzolanic activity of generated ash. Finally, the granular sewage sludge ash was milled in a ball mill (USSA/ball weight ratio of 0.10) during 50 minutes to increase its pozzolanic reactivity (Donatello et al. 2010; Pan et al. 2003a). This incineration process provided approximately 43 wt.% USSA regarding the sewage sludge mass incinerated.

Fig. 1. Process followed to obtain the uncontrolled-combusted sewage sludge ash.

Fig. 2. Temperature profile during the uncontrolled-combustion of the dried-granular sewage sludge.

USSA characterisation

The USSA was characterised by X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), Scanning electron microscopy (SEM), X-ray fluorescence (XRF), laser diffraction granulometry, insoluble residue according to UNE-EN 196-2:2014, BET surface area according to ISO 9277:2010, density

measured with pycnometer, and pH of 1g USSA to 10 ml deionized water, measured with pHmeter after 24 h. The XRD tests were run to 2θ range of 5–60°, using Cu-Kα radiation and a Ni filter at a voltage of 30 kV, a current intensity of 40 mA, an angle step of 0.02°, and a step time of 1.20 s/step. FTIR analyses were performed in the wavenumber range of 400 to 4000 cm^{-1}. SEM images using secondary electrons signal were obtained from the gold-covered surface of fractured pastes.

Compressive strength of PC/USSA mortars

Table 1 summarises the mix proportion and curing condition of the PC/USSA mortars developed. Percentages between 0-25 wt.% of PC were replaced by USSA, and the specimen which contained only PC (0 wt.% USSA) was prepared as a reference mortar. To all specimens, a constant water to cementitious materials ratio (w/cm) of 0.5, as well as a sand to cementitious materials (s/cm) ratio of 2 was used, considering the sum of PC and USSA as cementitious materials. The mixing procedure of the mortars was produced according to ABNT NBR 7215:2019, being the cementitious materials (PC and USSA) previously dry-mixed. The mortars were poured into cylindrical metallic moulds with 5 cm diameter and 10 cm height, as recommended by ABNT NBR 7125:2019, and they were compacted using a vibratory table for 1 minute. They were demoulded after 1 curing day, being maintained in high humidity (≈95%) and temperature-controlled chamber (25°C) until the compressive strength test. The compressive strength test was performed after 7, 28 and 90 curing days, using a universal testing machine with loading speed of 0,25 ± 0,05 MPa/s, in accordance with ABNT NBR 7215:2019.

Table 1. Mix proportion of the PC/USSA mortars.

Preparation and characterisation of the lime/USSA pastes (CH/USSA) and Portland cement/USSA pastes (PC/USSA)

CH/USSA pastes were prepared using a CH:USSA mass ratio of 3:7 (w/cm = 0.8) and 1:1 (w/cm = 1), where CH and USSA was taken into account as cementitious materials (cm). All these pastes were cured at 20 °C and 40 °C under high relative humidity conditions (RH > 95%). TG/DTG analyses were carried out at 1, 3, 7, and 28 curing days in the specimens cured at 40 °C, and at 3, 7, and 28 curing days in those

cured at 20 °C. The early TG/DTG test (1 day) for the pastes cured at 40 °C was performed to evaluate the acceleration of the pozzolanic reaction generated by temperature (Gastaldini et al. 2015).

PC/USSA pastes were prepared according to the mix proportions and curing conditions previously described in Table 1. TG/DTG and FTIR analyses were carried out in all the specimens, after 7, 28, and 90 curing days, to assess the microstructural development. The XRD tests were performed in the control paste and those containing 25 wt.% USSA, after being cured for 7, 28, and 90 days. SEM analyses were carried out only for the 0-USSA and 25-USSA pastes cured for 90 days.

The XRD, FTIR, and SEM analysis procedures were the same as those described in the USSA characterisation. An thermo-balance was used to analyse the pastes by thermogravimetry (TGA). The parameters employed for the TGA tests were as follows: temperature range, 35-600°C; heating rate, 10 °C.min^{-1}; and an atmosphere of N_2 (75 mL.min^{-1} flow). The samples were tested in sealed aluminium crucibles (100 µL) with a pinhole in the lid. Before those analyses, the pastes were grounded in an agate mortar, being the hydration process stopped with acetone as described by Moraes et al. (Moraes et al. 2016).

Results and Discussion

USSA characterisation

The physical characteristic and pH of USSA are summarized in Table 2. The USSA particle diameter size was significantly reduced after the milling process as can be seen in Fig. 3. The mean particle diameter of unmilled USSA was 199.41 µm, being composed of 50% (d(0.5)) of particles with a diameter under 95.19 µm (Table 2), likely due to an agglomerated particles aspect generated by the combustion of the dried-granular sewage sludge (Fig. 1). A large mean particle diameter for unmilled SSA was also reported by some authors (Donatello et al. 2010). The mean particle diameter of milled USSA was 20.28 µm, with d(0.1), d(0.5) and d(0.9) being 1.58 µm, 11.17 µm and 52.45 µm, respectively, as well as the volume of particles above 45 µm was 10.47%. The BET specific surface area of milled USSA was 14800 m^2/kg, which is a value close to the mean one found in the literature (15100 m^2/kg) (Cyr et al. 2007). This significant fineness could be the outcome of the particle size reduction during the milling process, which enhances the reactivity of the pozzolanic materials (Cordeiro and Kurtis 2017). Furthermore, the density of milled USSA was 2.05 g/cm^3, which agrees with the range (1.8 – 2.9 g/cm^3) reported in the literature to SSA (Lynn et al.

2015). The pH of milled USSA did not presented significant variation after 1 h and 24 h in deionized water (20°C), being the average value of 4.3. Such acid aspect could be the outcome of sewage sludge from anaerobic wastewater treatment which present a pH range of 3.57-6.43 (Hanum et al. 2019). As shown in Fig. 4, the milled USSA presented irregular shape, porous and rough particles, being similar to the morphologies reported by other authors (Chen and Poon 2017; Garcés et al. 2008; Naamane et al. 2016). These physical characteristics of milled USSA lead to a hydroscopic behaviour, and a reduction of the mortar workability, consequently, when it is used as a replacement for cementitious materials (Chang et al. 2020). The chemical composition of milled USSA are summarised in Table 3. As can be seen, the ash was mainly composed of 32.72 wt.% SiO_2, 20.72 wt.% Al_2O_3, and 11.27 wt.% Fe_2O_3. These values are similar to those previously reported by Chen and Poon (Chen and Poon 2017). As reported in the literature, these components of SSA chemical composition are the outcome of the type of wastewater treatment apart from the effluent sources. Al_2O_3 and Fe_2O_3 usually come from alum and ferric salts used during the wastewater treatment (Tantawy et al. 2012; UNESCO World Water Assessment Programme 2017). The quartz content (SiO2), in case of the USSA herein studied, likely came from the soil particles carried by the rain evacuation in the urban drainage system, which is jointly treated with the wastewater in the wastewater treatment plant. Furthermore, the presence of quartz in the SSA chemical composition, in some cases, could be the outcome of the quartz sand application during the wastewater treatment as nucleation sites for secondary iron minerals (Ma et al. 2018). As plotted in Fig. 5, the crystalline phases identified in USSA were quartz (SiO_2, PDFcard#331161), anhydrite ($CaSO_4$, PDFcard#371496) and hematite (Fe_2O_3, PDFcard#130534). It is well-known that the reactivity of a pozzolanic material highly depends on its amorphous content, which is denoted in the XRD pattern of USSA by a slight deviation of the baseline in the 18°–32° 2θ range (Moraes et al. 2015). In the studied sample, the intensity of the peaks attributed to quartz masks the deviation from the baseline. The milled USSA presented 27.20% of insoluble residues, which implied that a great amount of Al_2O_3 was amorphous, as well as a significant part of SiO_2, considering the low solubility of crystalline phases during to the insoluble residue test. The FTIR analyses performed on milled USSA are shown in Fig. 6. In agreement with the XRD results, the bands located at 1100, 1040, 671, 665, 611, and 455 cm^{-1} are attributed to Si-O-(Si, Al) vibrations (Criado et al. 2007; Tashima et al. 2017), and the Si-O double band at 796 – 778 cm^{-1} confirmed the presence of quartz (Criado et al. 2007).

Fig. 3. Granulometric distribution of milled USSA and unmilled USSA.

Table 2. Particle size, BET specific surface area, specific gravity and pH of USSA.

Table 3. Chemical composition of milled USSA (%, in mass).

Fig. 4. SEM micrographs of milled USSA: a) magnification of 1000x; b) magnification of 10000x.

Fig. 5. XRD pattern of milled USSA.

Fig. 6. FTIR of milled USSA.

Compressive strength development of the PC/USSA mortars

The compressive strength results of the mortars containing 0 to 25 wt.% USSA, cured at 25 °C for 7, 28, and 90 days, are reported in Fig. 7. As observed, increasing the USSA content generally improved the compressive strength, whatever the curing age. Similarly, for a given USSA content, the compressive strength was increased over time. Commonly, the literature have usually reported that the compressive strength of PC-based mortar is decreased as the PC replacement level by SSA is increased (Baeza-Brotons et al. 2014; Chen et al. 2013; Cyr et al. 2007; Lynn et al. 2015). However, some authors reported compressive strength of mortars made with a 10-20 wt.% SSA range in replacement of PC similar to one reached by a control mortar made with only PC (Chen and Poon 2017; Kappel et al. 2017). Chen and Poon (Chen and Poon 2017) observed that replacing up to 10 wt.% PC by SSA in mortars made with cementitious materials (PC + SSA), sand, water at a ratio of 1:2.75:0.484 did not reduce their compressive strength. Similarly, Kappel et al. (Kappel et al. 2017) reported comparable compressive strength values between mortars made with cementitious materials (PC + SSA), sand, water at a ratio of 1:3.0:0.5 replacing 20% PC by SSA and the reference mortar (only PC). Different compressive strength performance of the PC/SSA-based mortars reported in the literature are mainly due to the chemical composition of the SSA which could significantly vary depending on the sludge production and combustion method (Vouk et al. 2017) apart from the fineness that also affects its pozzolanic activity (Pan et al. 2003b). In the current study, the percentage of Al_2O_3 (20.72%) in USSA was superior to the average one (14.4%) reported in the literature (Lynn et al. 2015), that could explain the reasonable reactivity of the ash. The compressive strength values

of the mortars containing USSA cured for 90 days were in the range of 49.6-55.4 MPa, reaching values up to 11.5% higher than the one reached by the reference mortar (49.7 MPa after the same curing time).

The relative compressive strength gain (CSGr) was calculated according to Eq. 1, previously described by Monzó et al. (Monzó et al. 1999). This value was used to measure the compressive strength (in %) supplied by USSA to the mortars when compared with the hypothetical compressive strength given by an inert material (Monzó et al. 1999).

$$CSGr = \left[\frac{R_{C_i}}{R_{C_0}\, x\, w_C/(w_C+w_{USSA})} - 1\right] x\ 100 \quad (1)$$

Where R_{C_i} is the compressive strength of the USSA-containing mortar, R_{C_0} is the compressive strength of a reference mortar at the same curing age, w_C is the weight of cement, and w_{USSA} is the weight of USSA. The obtained CSGr results are plotted in Fig. 8. As observed, the CSGr increased as the USSA content increased, whatever the curing age (7, 28, and 90 days), reaching a higher value (69.8% for 25 wt.% SSA) at short curing time (7 days). Positive CSGr values were always obtained, which denotes that USSA clearly contributed to the development of mortar compressive strength. Results agree with those previously reported by Monzó et al. (Monzó et al. 1999), who also observed an improvement of CSGr with increasing SSA content.

Fig. 7. Compressive Strength of the PC/USSA mortar samples cured from 7 to 90 days.

Fig. 8. Relative compressive strength gain registered by the PC/USSA mortars containing 5 wt.% to 25 wt.% USSA, cured for 7, 28, and 90 days.

TG/DTG analyses of CH/USSA pastes

TG and DTG analyses were carried out on CH/USSA (3:7 and 1:1 mass ratio) pastes cured at 20 °C and 40 °C. Two distinct CH/USSA mass ratio and curing temperature conditions were evaluated to measure the extension of the pozzolanic reaction of USSA. Given that the consumption of the $Ca(OH)_2$ determines the

pozzolanic potential of USSA (Tironi et al. 2013), the $Ca(OH)_2$ fixed (CH_{Fixed}) by the ash was evaluated. To do so, the Eq.2, previously proposed by Payá et al. (Payá et al. 2002), was used:

$$CH_{Fixed}(\%) = \frac{CH_0 - CH_{USSA}}{CH_0} * 100 \quad (2)$$

where CH_0 and CH_{USSA} are the initial and final amounts of $Ca(OH)_2$, respectively, in the CH/USSA pastes.

The total mass loss and CH_{Fixed} values registered after the thermogravimetric analyses are reported in Table 4. The lowest amounts of fixed $Ca(OH)_2$ were registered at the shortest curing time (3 days, 42.5 % ± 0.5) with the 1:1 CH/USSA proportion. On the contrary, $Ca(OH)_2$ was totally consumed in the 3:7 CH/USSA system cured for 28 days (100% CH_{Fixed}). Besides, in the system with a CH/USSA mass ratio of 1:1, the maximum content of CH_{Fixed} at 20 °C and 40 °C was 61.4 % and 86.1 %, respectively. The obtained results confirmed the expected pozzolanic behaviour of USSA, given its fineness and chemical composition, previously described in the USSA characterisation section.

Three different regions were identified in the DTG curves of the CH/USSA pastes, which are plotted in Fig. 9. The first region R_1, from 100°C to 180°C, was associated with the mass loss due to the dehydration of calcium silicate hydrates (C-S-H) and ettringite ($C_3A.3CaSO_4.32H_2O$ – Aft) (Payá et al. 2002). The second region R_2, from 180°C to 300°C, was attributed to the mass loss originated by the dehydration of calcium silicate aluminate hydrates (C-A-S-H) and calcium aluminate hydrates (C-A-H) (Payá et al. 2002; Shatat 2016). Finally, the third region R_3, from 520°C to 600°C, was assigned to dehydration of the $Ca(OH)_2$ (Soriano et al. 2013).

Table 4. Mass loss registered after the TG/DTG analyses of the CH/USSA pastes (R_1, C-S-H and Aft; R_2, C-A-S-H and C-A-H; R_3, $Ca(OH)_2$ dehydration) and the calculated percentage of fixed $Ca(OH)_2$ (CH_{Fixed}).

As Fig. 9 shows, the band arising from 520°C to 600°C disappeared in the CH/USSA 3:7 pastes after 28 curing days at 20°C or 3 curing days at 40°C. This behaviour was explained by the consumption of $Ca(OH)_2$ due to the pozzolanic reaction (Moraes et al. 2015; Payá et al. 2002; Soriano et al. 2013). The peak in the R_3 region disappeared earlier in the specimen cured at 40°C than in that under 20°C which, as pointed out by Mirzahosseini and Riding (Mirzahosseini and Riding 2014), occurred because higher temperatures

accelerate the pozzolanic reaction. The $Ca(OH)_2$ dehydration band appeared in all of the CH/USSA 1:1 specimens. Although its intensity reduced with higher temperatures or longer curing times, its presence indicated that higher amounts of USSA were required to consume all the $Ca(OH)_2$ in the CH/USSA 1:1 pastes.

As previously reported in Table 3, USSA contained a high percentage of Al_2O_3 (20.72%), most probably due to the presence of different types of phyllosilicates in the sewage sludge. During the combustion process, these phyllosilicates decompose providing amorphous alumina that may react with $Ca(OH)_2$ and reactive silica (also present in USSA) to produce aluminium hydrates. This would explain the broad band in the R_2 region of the DTG curves, originated by the dehydration of C-A-H and C-A-S-H.

Fig. 9. DTG curves for the CH/USSA pastes prepared with a mass ratio of 3:7 and 1:1, cured at 20 and 40°C for 1, 3, 7, and 28 days.

TG/DTG analyses of PC/USSA pastes

TG/DTG analyses were performed on PC/USSA pastes containing up to 25 wt.% USSA, and the results are summarised in Table 5 and Fig. 10. To assess the pozzolanic reaction of these pastes, the CH_{Fixed} was also calculated, according to the Eq. 3 proposed by Soriano et al. (Soriano et al. 2013).

$$CH_{Fixed}(\%) = \frac{(CH_c xC\%) - CH_{USSA})}{(CH_c xC\%)} * 100 \quad (3)$$

Where CH_c was the amount of $Ca(OH)_2$ in the reference paste (0-USSA), CH_{SSA} was the amount of $Ca(OH)_2$ in the PC/USSA pastes and $C\%$ was the proportion of PC in the mix.

As reported in Table 5, the 5-USSA (5 wt.% USSA) paste cured for 7 days presented a negative CH_{Fixed} value. This is explained by the further hydration of the PC due to the significant amount of fine particles (d(0.5)=11.17 µm, Fig. 3) of USSA, that displayed nucleation site role, then yielding higher amount of available $Ca(OH)_2$ (Jaturapitakkul et al. 2011; Khan et al. 2017; Soriano et al. 2016). Besides, the content of amorphous aluminosilicate phases provided by USSA in the 5-USSA specimen could be insufficient to consume a significant percentage of $Ca(OH)_2$ produced in the Portland cement hydration. However, in the pastes prepared with the highest amount of USSA (25-USSA, 25 wt.% USSA), the content

of amorphous aluminosilicate phases supplied by USSA was noteworthy and, therefore, the pozzolanic effect was probably superior to the particle effect. This hypothesis was corroborated by the CH_{Fixed} values, since they reached a maximum of approximately 80 % in the paste prepared with 25 wt.% USSA cured for both 7 and 90 days (25-USSA). Similar results were previously reported by Baeza-Brotons et al. (Baeza-Brotons et al. 2014), who also observed a progressive increase of the fixed $Ca(OH)_2$ with increasing SSA contents, and reported a value of 33.28 % when replacing 20 wt.% of PC by SSA.

For a given USSA content, the CH_{Fixed} value oscillated with the curing time. The hypothesis for such a phenomenon is the combination of the pozzolanic effect, which consumes $Ca(OH)_2$, with the particle effect, which accelerates the PC hydration and thus, generates more $Ca(OH)_2$. After 90 curing days, when the hydration of the Portland cement seems to be stable, the specimens with the highest USSA content consumed the highest amounts of $Ca(OH)_2$.

The DTG curves of the PC/USSA pastes were also divided into three main regions, depending on the registered dehydration bands (R_1, R_2 and R_3). The mass loss in region R_1 was linked to the formation of the C-S-H gel and ettringite, while the bands appearing in the R_2 area denoted the dehydration of C-A-S-H and C-A-H. These products typically form after the hydration of PC and pozzolanic reactions (El-Diadamony et al. 2018; Jeon et al. 2018; Mastali et al. 2018). According to the DTG results, the mass loss in region R_2 increased with the curing time and PC substitution, which is attributed to the reactivity of the alumina contained in USSA. The slight signal arising at 417 °C could originate from the dehydration of brucite ($Mg(OH)_2$), from the reactive magnesia (MgO) present in USSA or PC (Imbabi et al. 2012; Zhang et al. 2015). In agreement with the fixed $Ca(OH)_2$ results, the mass loss in the region R_3 decreased with increasing USSA contents, which confirms that the $Ca(OH)_2$ produced in the hydration of Portland cement was consumed during the pozzolanic reactions of USSA. The TG/DTG results are in line with the compressive strength evolution of the PC/USSA mortars shown in Fig. 7 since the mechanical properties also improved with increasing ash contents or longer curing times.

Fig. 10. DTG curves of PC/USSA pastes prepared with 100 wt.% PC (0-USSA) and 5-25 wt.% USSA (5-USSA, 15-USSA, 25-USSA), cured at 25 ºC for 7, 28, and 90 days.

Table 5. Mass loss and percentage of fixed $Ca(OH)_2$ (CH_{Fixed}) registered during the TG/DTG tests of PC/USSA pastes.

FTIR analyses of PC/USSA pastes

All PC/USSA pastes presented similar FTIR spectra, and the results are shown in Fig. 11. The band at 3639 cm^{-1} was assigned to the stretching vibrations of the structural O-H group in $Ca(OH)_2$ (Moraes et al. 2015). In consonance with the TG/DTG results, this band tended to disappear with higher amounts of USSA or longer curing times, which corroborates the occurrence of the pozzolanic reaction. The bands at 3392 cm^{-1} and 1641 cm^{-1} were assigned to the stretching and bending vibration, respectively, of the O-H group in the calcium aluminosilicate hydrate (C-A-S-H), generated by the hydration of PC and the pozzolanic reaction (Biricik and Sarier 2014; Kapeluszna et al. 2017; Kumar et al. 2018). The asymmetric stretching vibration of the Si-O-T (T=Si, Al) from the C-S-H and C-A-S-H gels appeared at 958 cm^{-1} (Kapeluszna et al. 2017). All of the spectra presented transmittance bands located at 1412 cm^{-1} and 874 cm^{-1}, which were attributed to the asymmetric and stretching vibrations of the C-O bonds in $CaCO_3$ (Tantawy 2017). The signal at 1091 cm^{-1} was linked to the stretching vibration of the S-O bonds (Kumar et al. 2018; Tantawy 2017). This band also arose in all the specimens, mainly at early curing ages, and corroborated the presence of gypsum and the formation of ettringite during the PC hydration.

Fig. 11. FTIR spectra of the PC/USSA pastes prepared with 0 wt.% USSA (a) 5 wt.% USSA (b), 10 wt.% USSA (c) and 25 wt.% USSA (d); all of them cured at 25 °C for 7, 28, and 90 days.

XRD analyses of PC/USSA pastes

The XRD analyses were run on the reference paste (0-USSA) and for that containing 25 wt.% USSA (25-USSA), which presented the highest compressive strength (Fig. 7) and fixed $Ca(OH)_2$ values (Table 6). The XRD patterns are presented in Fig. 12. As observed, signals due to the formation of ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12}.26H_2O$, PDFCard#00411451) arose in both spectra (0-USSA and 25-USSA), mainly at short curing ages. Peaks associated with the presence of monosulfate ($Ca_4Al_2SO_{10}.12H_2O$, PDFCard#180275) were also distinguished after 90 curing days. These could have resulted from the

366 transformation of ettringite or have directly formed from the reaction of $Ca_3Al_2O_6$ (C_3A) in the presence of
367 small amounts of gypsum (Christensen et al. 2004). The peaks attributed to calcite ($CaCO_3$,
368 PDFCard#050586), which arose in both samples, were associated with its presence in PC or slight
369 carbonation of the pastes.

370 The brucite ($Mg(OH)_2$, PDFCard#16747) and gypsum ($CaSO_4$, PDFcard#371496) peaks identified in the
371 25-USSA XRD pattern might be due to the presence of MgO and SO_3 in the original USSA (Table 3).
372 Signals originated by carboaluminate phases ($Ca_4Al_2O_6CO_3.11H_2O$, PDFCard#410219) also arose in both
373 specimens, 0-USSA and 25-USSA, most probably resulting from the reaction between anhydrous calcium
374 aluminate and $CaCO_3$ (Segui et al. 2012). The main portlandite peaks ($Ca(OH)_2$, PDFCard#040733) in the
375 XRD pattern of the 25-USSA paste decreased over time, confirming the consumption of $Ca(OH)_2$ due to
376 the USSA pozzolanic reactions. Furthermore, a broader diffusive halo was observed in the XRD pattern of
377 the 25-USSA paste over time, which means a larger amount of amorphous hydrated phases over time,
378 endorsing the occurrence of the pozzolanic reaction. The broad diffusive halo observed in the XRD patterns,
379 for a given curing age, was more noteworthy in the 25-USSA paste than in the reference sample, which
380 confirmed the presence of a higher amount of amorphous phases after partially replacing PC by USSA, and
381 thus a greater compressive strength of the sample 25-USSA, in line with the compressive strength results.

382

383 **Fig. 12.** XRD spectra of the reference paste (0 USSA - black line) and the PC/USSA paste prepared with
384 25 wt.% USSA (25 USSA - red line); samples were cured at room temperature for (a) 7 days, (b) 28 days,
385 and (c) 90 days.

386 **SEM analyses**

387 The SEM analyses were conducted on 0 USSA and 25 USSA pastes cured for 28 and 90 days. As shown
388 in Fig. 13, all samples exhibited a dense microstructure with similar reactions products, such as hydrated
389 gehlenite, C-A-S-H, C-S-H or ettringite. All these products were previously identified by TG/DTG, FTIR
390 or XRD analyses and typically formed during the cement hydration or pozzolanic reaction.

391

392 **Fig. 13.** SEM micrographs of the 0-USSA paste cured for 28 (a) and 90 days (b), and the 25-USSA paste
393 cured for 28 (c) and 90 days (d). Ettringite (ET), hydrated gehlenite (GEH), and C-S-H gels (C-S-H).

Conclusion

A simple and economic uncontrolled-combustion process was used to produce sewage sludge ash (USSA). The reactivity of this ash was investigated, with the following results:

- The chemical composition of USSA was similar to that reported in the literature for the SSA obtained from controlled-combustion processes.

- The USSA exhibited a high Al_2O_3 content (20.72 wt.%), which was attributed to the presence of phyllosilicates in the sewage sludge, that yielded amorphous alumina after their thermal decomposition.

- The pozzolanic reaction of USSA with $Ca(OH)_2$ liberated during the hydration of PC originated hydrated compounds (C-S-H, C-A-S-H and C-A-H). The formation of hydrated products enhances the mechanical strength development of the mortars.

- Mortars containing 25% of SSA yielded increments of 27%, 16% and 7% after 7, 28 and 90 curing days, respectively.

- A maximum relative compressive strength gain of 69.8 % was registered, which was provided by the mortar prepared with 25 wt.% USSA, cured at 25 ºC for 7 days.

This research adds knowledge to the existing studies, which generally used sewage sludge ash produced under temperature and time-controlled processes, in technological incineration plants. The novelty is based on: a) the uncontrolled-combustion of the sewage sludge can generate ash with a low loss on ignition; and b) the obtained ash presents good pozzolanic activity, improving significantly the mechanical development of Portland cement-based mortar when used as supplementary cementing material. This study may encourage further investigations, aiming to promote new solutions to manage the waste generated in wastewater treatment plants which, due to economic and technological issues, is currently being deposited mainly in landfills.

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Data Availability

Some or all data, models, or code that support the findings of this study are available from the corresponding author upon reasonable request.

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629

630 **Table 1.** Mix proportion of the PC/USSA mortars.

Mortar samples	PC	USSA	w/cm	s/cm	Curing Environment
	% mass		(mass ratio)		
0-USSA	100	0	0.5	2	Moisture room (relative humidity≈ 95%, 25°C)
5-USSA	95	5			
10-USSA	90	10			
15-USSA	85	15			
20-USSA	80	20			
25-USSA	75	25			

631

632 **Table 2.** Particle size, BET specific surface area, specific gravity and pH of USSA.

	Milled USSA	Unmilled USSA
Mean diameter	20.28 μm	199.41 μm
d(0.1)	1.58 μm	4.43 μm
d(0.5)	11.17 μm	95.19 μm
d(0.9)	52.45 μm	539.70 μm
Particle above 45 μm	10.47 %	-
BET specific surface area	14800 m^2/kg	-
density	2.05 g/cm^3	-
pH	4.13	-

Table 3. Chemical composition of milled USSA (%, in mass).

Major Oxides	SiO_2	Al_2O_3	Fe_2O_3	P_2O_5	CaO	SO_3	TiO_2	MgO	K_2O	Na_2O	Others	LOI
USSA	38.28	20.72	11.27	7.28	5.51	4.18	3.73	1.91	0.73	0.70	1.97	3.72

Table 4. Mass loss registered after the TG/DTG analyses of the CH/USSA pastes (R_1, C-S-H and Aft; R_2, C-A-S-H and C-A-H; R_3, $Ca(OH)_2$ dehydration) and the calculated percentage of fixed $Ca(OH)_2$ (CH_{Fixed}).

CH/USSA, mass ratio	T, °C	Curing age, days	Mass loss (%)			Total mass loss (%) (35-600°C)	CH_{Fixed} (%)
			R_1 (100-180°C)	R_2 (180-300°C)	R_3 (520-600°C)		
3:7	20	3	1.5	5.4	2.0	11.3	70.6
		7	1.9	5.9	1.5	11.7	78.7
		28	3.0	8.4	-	14.4	100
	40	1	1.7	4.5	2.2	11.0	68.1
		3	2.2	5.5	0.1	11.4	98.4
		7	2.8	6.5	_	12.4	100
		28	3.4	6.3	_	13.3	100
1:1	20	3	1.5	4.6	6.6	14.7	42.5
		7	1.9	4.9	6.4	15.5	44.4
		28	2.3	7.0	4.4	17.2	61.4
	40	1	1.5	4.8	6.5	14.8	43.5
		3	2.2	5.5	4.9	15.2	57.6
		7	2.8	5.9	3.6	15.8	68.7
		28	2.5	5.9	1.6	14.5	86.1

Table 5. Mass loss and percentage of fixed $Ca(OH)_2$ (CH_{Fixed}) registered during the TG/DTG tests of PC/USSA pastes.

Curing days	Specimens	Mass loss (%)			Total loss (%) (35-600 °C)	CH_{Fixed} (%)
		R_1 (100-180 °C)	R_2 (180-300 °C)	R_3 (520-600 °C)		
7	0-USSA	6.8	3.6	1.7	16.0	-

	5-USSA	8.1	3.9	1.7	18.1	-9.2
	15-USSA	8.6	4.4	0.8	18.1	44.2
	25-USSA	8.7	4.2	0.2	17.7	80.2
28	0-USSA	6.3	3.5	1.2	14.7	-
	5-USSA	8.1	3.8	1.0	16.9	9.0
	15-USSA	7.5	4.0	0.4	15.9	60.8
	25-USSA	10.6	4.9	0.4	21.1	58.9
90	0-USSA	5.2	4.0	1.9	15.7	-
	5-USSA	5.8	4.9	1.7	17.5	5.8
	15-USSA	5.1	5.9	1.0	18.0	38.9
	25-USSA	6.1	6.1	0.3	17.7	79.8

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