Postprint of: Świerczek L., Cieślik B., Konieczka P., Challenges and opportunities related to the use of sewage sludge ash in cement-based building materials – A review, JOURNAL OF CLEANER PRODUCTION, Vol. 287 (2021), 125054, DOI: 10.1016/j.jclepro.2020.125054

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# 1. Introduction

Sewage sludge (SS), produced in every sewage sludge treatment plant, can become a significant source of energy, heat, or chemical substances e.g. fertilizers, macro- and microelements. Due to the presence of mentioned nutrients, in many countries, SS constitutes a valuable substrate for the improvement of arable land quality or remediation of degraded areas, which is a cost-effective and environmentally friendly solution. Annually, around 11.5 Mt of SS dry matter (DM) is produced in European Union countries (Vouk et al., 2018). It is estimated that the amount of SS produced in these countries by 2020 will have increased to about 13 Mt DM (D. Vouk et al., 2017). The available SS disposal methods are limited by a series of constantly updated legal acts (Cieślik et al., 2015). For this reason, existing methods are being replaced in favour of more modern ones, that are consistent with new legal requirements. One of the legal act in the European Union regarding the SS is Council Directive (91/271/EEC) of 21 May 1991 (European Commission, Council Directive of 21 May, 1991) according to which the SS should be treated as a product for further development. For this reason, some of the existing management techniques (e.g. storage, soil reclamation, fertilizer production) are even forbidden (Kacprzak et al., 2017).

Sewage sludge management methods can be divided into two main groups: biological and thermal. Biological methods such as composting or anaerobic digestion are used less frequently due to the increasing legal requirements for the quality of the processed SS which usually requires further processing (Cieślik et al., 2015). Thermal methods that become a promising alternative include the incineration, pyrolysis and vitrification. The treatment of SS by pyrolysis generates less pollution than incineration, but the main barrier is the complexity of the process, which generates significant maintenance costs. Vitrification, belonging to the high-temperature process (1000-1600 °C), ensures the production of completely environment-neutral material. However, due to high energy consumption, it also requires high financial expenditures (Samolada and Zabaniotou, 2014). In addition, during the vitrification, specific elements species which could be described as volatile in certain conditions (As, Cd, Pb, Se, Hg) (Elled et al., 2007), can be released into the environment so there is a need for continuous

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monitoring of the process. Moreover, the variety of by-products streams, which have to be managed, also should be considered (Hernandez et al., 2011).

Due to the diverse composition of SS and significant amounts of potentially harmful substances contained in it (drugs, pharmaceuticals (Díaz-Cruz et al., 2009; Nieto et al., 2010)) and heavy metals (Lu et al., 2016; Yang et al., 2017)), this waste is usually utilized by incineration. The process ensures complete decomposition of pathogens, organic matter, and odoriferous substances. Additionally, the volume of SS is significantly reduced, even by 90 - 95%, and its mass by about 70% compared to the state before the process (Lewkowska et al., 2016; Lynn et al., 2015; Tashima et al., 2017). The disadvantage of the incineration process is the concentration of potentially harmful elements and unavoided production of solid residues (dust and ash), which must be properly managed.

Positive aspects related to the incineration process make it an increasingly common management technique. Around 22% of SS in the European Union is incinerated (D. Vouk et al., 2017) which gives about 0.77 Mt solid waste per year. Germany itself produces about 0.3 Mt of SSA (Krüger and Adam, 2014). It was estimated that the amount of SSA generated in the USA is about 0.54 Mt (Cyr et al., 2007) while in Japan, the management problem is related to significant quantities of obtained SSA (about 0.24 Mt per year) and limited land availability (Donatello and Cheeseman, 2013). It is worth noting that the presented values refer only to the amount of the produced ash, i.e. one of the solid residue fractions produced during the thermal utilization process. As a result of the exhaust gas stream purification, because of the use of various sorbents (Kijo-Kleczkowska et al., 2015; Liu et al., 2015) a second waste fraction is also formed – dust. The dust to ash ratio may vary from 5% to 60%, depending on the technology (Lapa et al., 2007; Marani et al., 2003), and in most publications, related to SSA management, the problem of dust management is usually completely neglected.

The quantities of generated SSA in comparison to ash from the energy industry (coal combustion residuals) are about 100 times smaller (about 115 Mt for UE and US countries) (Belviso, 2018; Cyr et al., 2007). However, the management of significant amounts of coal combustion residuals is not considered as a problem, because it is a commonly used mineral addition in cementitious binders. Due to many similarities in SSA production and similar composition to coal fly ash, research on the use of SSA in the construction industry is often carried out. Sewage sludge ash is treated often as a replacement for a binder (Chen et al., 2018; Kappel et al., 2017; Dražen Vouk et al., 2017) or as a mineral addition (Baeza-Brotons et al., 2014; Jamshidi et al., 2012; Záleská et al., 2018) to both mortars and concretes. Mentioned waste can be also used in alkali-activated binder mixtures as a reaction precursor.

(Chakraborty et al., 2017; Istuque et al., 2016; Tashima et al., 2017). Bricks and tiles (Lin et al., 2017, 2016; Mozo and Gomez, 2016), ceramics (Zhang et al., 2015), glassy materials (Borowski, 2015; Celary and Sobik-Szołtysek, 2014) and lightweight aggregates (Cheeseman and Virdi, 2005; Chiou et al., 2006; Lin, 2006) belonging to sintered materials can be also produced with the contribution of SSA, however, these processes also require significant energy demand. The manufactured products, despite eliminating the risk of heavy metals entering the environment, are often characterized by inferior properties e.g. lower compressive strength or higher shrinkage. In addition, the production of such materials requires additional gas treatment systems due to the possible presence of volatile heavy metals and other elements species.

Due to the variety of research presented in publications, the complexity of the used waste and the various conclusions regarding the impact of SSA on the properties of cement-based construction products, this article summarizes and presents the latest literature reports related to the use of SSA in cementitious materials. Considering the disadvantages of selected development techniques of both SS and SSA and the fact that SS incineration is currently considered as the Best Available Technology (Kijo-Kleczkowska et al., 2016), the article focuses exclusively on the management of SSA in concretes and mortars based on hydraulic binders. This approach eliminates the possible emission of heavy metals to the environment (which is confirmed by leaching tests) and does not require complicated facilities. However, it is difficult to estimate the cost and profitability of such management technique, because it depends on local situation and circumstances, such as raw materials availability, economy or geography (Spinosa et al., 2011).

Literature reports have been described in detail in two main groups: cementitious materials (mortars or concretes), in which SSA is an additive and materials in which SSA is a substitute for the main binder. It was also indicated which of the factors may affect the properties of the end products e.g. the proportion and type of binder, the method of sample preparation as well as the impact of SSA quality on durability properties. Due to the difficulty in clearly determining whether toxic heavy metals contained in SSA are safely immobilized in a cement matrix, results of leaching heavy metals from hardened products are also described. The presented information can be particularly helpful during the design of modern building materials and the analysis of the environmental impact of these materials. Some critical issues are also considered.

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# 1. Methods and scope

This study focuses on describing SSA management methods and presents the advantages and disadvantages of including mentioned wastes into building materials based on cement binders. A thorough assessment of selected approaches available in the literature has been carried out. Since the subject is still developing in some countries, the review is based on literature from the entire world. The article is based on the scientific literature but not exclusively. Research published in technical journals and book sections was also used.

The article briefly presents the variety of research on the use of SSA in cement building materials, the research on the cement hardening process, the impact of mineral additives on the properties of mortars and concretes, as well as the use of other waste materials in the construction industry. A thorough analysis of the influence of various factors on cement building materials was performed to simplify condensed information on the impact of SSA wastes uses in building materials.

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# 2. The SSA potential in materials based on cement binders

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# 2.1. Characteristics of SSA

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Sewage sludge is a residue left after a primary (physical), secondary (biological) and tertiary (chemical) wastewater treatment (Fytili and Zabaniotou, 2008). Some of the produced residues, especially in municipal wastewater treatment plants processing significant amounts of wastewater, are processed by anaerobic digestion. Before incineration, the produced SS is dewatered and dried in order to remove as much water as possible. If the SS calorific value is low (e.g. due to high water content), additional fuel can be used in the incineration process (Wzorek and Tańczuk, 2015). The properly prepared SS is most commonly incinerated in fluidized bed furnaces at a temperature of 850 °C in order to utilize potentially harmful organic fractions. The process can be carried out at higher temperatures to degrade e.g. polychlorinated substances (Wey et al., 2008). Exhaust gases are dedusted in a multi-stage cyclone system, bag filters or electrostatic precipitators, which results in the separation of ash (fly ash). Further gas purification with the use of sorbents, for example, sodium bicarbonate, lime and activated carbon (used for neutralization and removal of harmful organic substances and volatile elemental fractions) generate another fraction of waste – dust or fine fly ash. Both streams of solid residues due to the presence of heavy metals and metalloids have to be properly managed (Kacprzak et al., 2017; Samolada and Zabaniotou, 2014). An example diagram of a sewage

sludge incineration system is presented in Figure 1. It is worth noting that the type of used sorption media may cause differences in the composition of dust obtained during exhaust gas purification. The elemental composition of dehydrated SS and SSA are also significantly different, therefore ash fraction constitutes a completely different product (Świerczek and Cieślik, 2018).

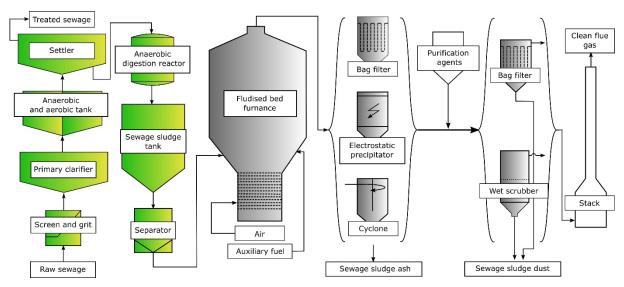


Figure 1. A simplified diagram of a SS thermal treatment plant.

The main oxides found in SSA, determined mostly using X-ray fluorescence technique (XRF) include SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, CaO, Fe<sub>2</sub>O<sub>3</sub>, NaO, MgO and P<sub>2</sub>O<sub>5</sub> (Lynn et al., 2015). Silicone oxide minerals (e.g. quartz), whitlockite, mullite, calcium sulphate minerals (e.g. anhydrite), magnetite, feldspars, and micas belong to the crystalline minerals (shown in Fig. 3) present in SSA. However, the amorphous phase constitutes about 45 % of the SSA mass (Chen and Poon, 2017; Cyr et al., 2007; Halliday et al., 2012). In addition to the aforementioned main oxides, SSA contains a number of inorganic heavy metal compounds. Based on the analysis of many test results, it has been shown that solid residues contain Cd, Cu, Ni, Pb, Cr, Mn, Fe, As, Sb, Hg at various concentrations. Reduction of the SS volume and mass as a result of incineration increases the content of heavy metals in the obtained solid residues (Van de Velden et al., 2008).

#### 2.2. Possibilities and limitations related to the use of SSA in cement-based materials

The main silicate compounds in cement clinker that are responsible for its hardening are alite (tricalcium silicate  $3CaO\cdot SiO_2$ ) and belite ( $\beta$ -dicalcium silicate  $-2CaO\cdot SiO_2$ ). Alite constitutes about 50%, while belite about 20% of the total mass of clinker. The remainder of the mass are mostly tricalcium aluminate ( $3CaO\cdot Al_2O_3$ ), and tetracalcium aluminoferrite



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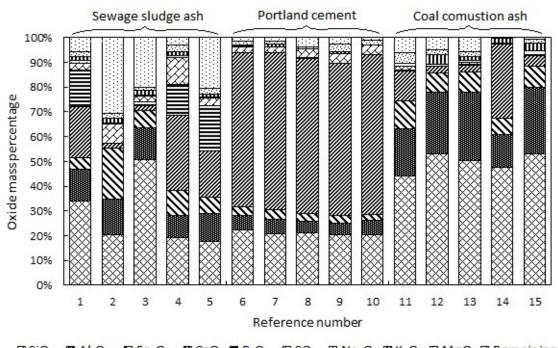
(4CaO·A1<sub>2</sub>O3·Fe<sub>2</sub>O<sub>3</sub>) (Li et al., 2018; MacKenzie and Smith, 2002). As a result of the reaction of these substances with water, calcium silicate hydrate and calcium hydroxide are formed. The hydrated calcium silicates commonly described as C-S-H phase (where: C-CaO, S-SiO<sub>2</sub>, H-H<sub>2</sub>O) may have a various composition, morphology, and degree of crystallization. The introduction of e.g. supplementary cementitious materials (SCM) e.g., fly ash, slags, calcined clays containing aluminium causes the formation of calcium aluminosilicate hydrate C-A-S-H (where: A-Al<sub>2</sub>O<sub>3</sub>) (Geng et al., 2017; Li et al., 2019). Both mentioned phases are responsible for the strength of the cement paste. However, some minerals from the calcium aluminate hydration system (e.g. CAH<sub>10</sub>, C<sub>3</sub>AH<sub>6</sub>) have been shown to have even higher mechanical properties than the C-S-H phases. Despite the low density of C-A-S-H minerals, similar or higher mechanical properties are mainly related to the topology of the mineral network (Geng et al., 2018).

It should be noted that the use of SCM is becoming a more and more common procedure aimed at reducing CO<sub>2</sub> emissions and cement consumption. Due to the use of new types of SCM or their blends, the methods for determining technical standards and specific criteria for determining their suitability in building materials also need to be modified/updated. In accordance with ASTM C 618-19, pozzolanic material that can be used in concrete should contain SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and Fe<sub>2</sub>O<sub>3</sub> in an amount of 50 - 70% of the total mass of the material. Additionally, such materials generally do not exhibit binding properties, but form stable bonds in the presence of water and Ca(OH)<sub>2</sub> (ASTM, 2003). However, as emphasized by R. Snellings methods for determining SCM activity based on the measurement of the lime consumption or measuring only the strength of the SCM - cement mixture are insufficient. The authors propose the measurement of cumulated heat release, chemical shrinkage, or bound water content, which allows treating a wider range of SCMs as active materials in cement mixtures. (Snellings et al., 2019). The differences in the methods of determining SCM activity in cement mixtures may be related to the lack of uniformity regarding the activity of SSA in research on its use in building materials.

The physicochemical characteristics, composition and production technology of SSA, make it similar to the coal ash residuals thus SSA is used as a binder or fine aggregates substitute which has its advantages and disadvantages. The presence of active oxides in SSA, mainly SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, CaO can have a positive effect on the hardening of building materials (Chen and Poon, 2017; Sara Naamane et al., 2016). Figure 2 presents a comparison of the main oxides percentage content found in SSA, Portland cement and coal fly ash. The coal fly ash, similar to SSA, includes more Al<sub>2</sub>O<sub>3</sub> compared to Portland cement. The presence of this oxide,

as well as sulfate and calcium ions, contribute to the formation of calcium sulfoaluminate hydrates (AFt) e.g. ettringite and calcium aluminate hydrates (AFm) e.g. monosulfoaluminate (Avet et al., 2019; Balonis, 2019). The AFm phase is characterized by significant chloride sorption potential, therefore the presence of SSA in concrete may increase its resistance to the harmful effects of chlorides (Balonis, 2019; Lynn et al., 2015). It is worth noting that SSA in mortar or concrete can also increase the number of nucleation sites for C-S-H phase formation (Scrivener et al., 2015).

An additional benefit associated with the use of SSA is the possibility of immobilizing heavy metals in the matrix of hydrated phases (K.Kazberuk, 2011; Li et al., 2017). Due to the characteristic particle-size distribution of SSA (2.5 - 250.0 µm), it can be used as a filler or substitute for fine aggregate in concrete (Lynn et al., 2015).



☑ SiO<sub>2</sub> ■ Al<sub>2</sub>O<sub>3</sub> ☑ Fe<sub>2</sub>O<sub>3</sub> ☑ CaO ■ P<sub>2</sub>O<sub>5</sub> ☑ SO<sub>3</sub> □ Na<sub>2</sub>O Ⅲ K<sub>2</sub>O ☑ MgO ☑ Remaining

Figure 2. Mass percentage of main oxides in SSA, Portland cement and coal combustion ash; 1 -(Cyr et al., 2007), 2 - (Tay and Show, 1994), 3 - (Pan et al., 2003), 4 - (Garcés et al., 2008), 5 - Rutkowska et al., 2018), 6 - (Li and Poon, 2017), 7 - (Tay and Show, 1994), 8 - (Monzo et al., 1997), 9 - (Yusuf et al., 2012), 10 - (Monzó et al., 1996), 11 - (Chen and Poon, 2017), 12 - (Fernández-Jiménez and Palomo, 2005), 13 - (Jang and Lee, 2016), 14 - (Suksiripattanapong et al., 2015), 15 - (Richardson et al., 2016).

Despite the similar ratio of  $SiO_2$  /  $Al_2O_3$  to coal fly ash, the sum of oxides in SSA on the basis of which material can be classified as pozzolan material is often lower (Fig. 2). It was reported that SSA exhibit lower pozzolanic activity than ash from coal combustion (Chen and Poon, 2017; Rutkowska et al., 2020) or other conventional supplementary cementitious material

(Mejdi et al., 2020). Figure 3 presents the differences in mineralogical composition between SSA and coal fly ash. The higher ash activity compared to SSA is associated with a different composition and the occurrence of an amorphous phase. It is worth adding that the SSA's mineralogical composition is primarily influenced by the processing temperature of the SS. That basically means that it is impossible to substitute fly ash with SSA without at least a slight change in product properties.

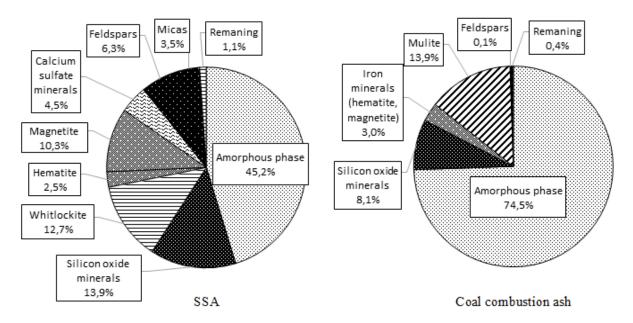


Figure 3. Comparison of the content of the main mineral phases determined by the XRF technique in SSA and ash from coal combustion (Chen and Poon, 2017; Cyr et al., 2007; Hadi et al., 2018; Halliday et al., 2012; Jang and Lee, 2016; Mádai et al., 2015; Rentsenorov et al., 2018; Schöler et al., 2015; Yan et al., 2018; Záleská et al., 2018).

Unfortunately, some of the researches are based on the use of SSA obtained in the laboratory (S Naamane et al., 2016; Dražen Vouk et al., 2017), while other use SSA which is taken from the SS incineration facilities (Chen and Poon, 2017; Kappel et al., 2017; Li et al., 2017; Piasta and Lukawska, 2016). Similar elements present in ash and dust obtained during technological scale incineration makes it impossible to compare with SSA incinerated in the laboratory furnace. It should be remembered that when SS is processed in incineration plants, it could be pre-stabilized by adding lime. The SSA obtained from lime-stabilized SS can be much more active than SSA prepared in the laboratory (Mejdi et al., 2020). Moreover, the technological processing of SS is different from the laboratory processing because some of the analytes are lost during the SS firing in the laboratory furnace (Tang et al., 2008; Youcai et al., 2004).

In addition, the laboratory production of SSA causes differences in the mineralogical composition, therefore, a sample of SSA produced on a simple, laboratory procedure is unrepresentative which may cause significant differences, e.g. instability or differences in durability, between cementitious materials containing technological or laboratory ash.

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### 3. Sewage sludge ash in mortars as a partial binder or aggregate substitute

Sewage sludge ash can be treated as a substitute for the main binder or an additive in the mortar. The purpose of partial binder replacement is mainly to reduce its demand. The second type of research considered in the review paper is the use of SSA as an additive with and without removal of aggregate. Despite the differences in the description of the methodology itself, the presented approaches are similar. The average binder content based on the fresh weight of the mortar is 20% while SSA amounts 4.5% (Fig. 4). In general, partial substitution of the main binder or aggregate on SSA causes deterioration of mortar durability even after long curing time (Chang et al., 2010; Chen and Poon, 2017; Piasta and Lukawska, 2016). Table 1 presents the impact of SSA on the basic parameters of mortars with its addition. The deterioration of the strength properties is related to the lack of pozzolan reaction between the binder components as well as reduction of the total binder mass content in the mixture (Chen and Poon, 2017; Li et al., 2017; S Naamane et al., 2016).

Table 1. Characteristic of mortar samples containing SSA. 262

Type of	The share of	Type of aggregate	W	S	WA	CS	Reference
cement	SSA* [%]						
OPC	2.9	Glass <5mm	7	n.d.	7	7	(Li et al., 2017)
OPC	4.7	Sand +	7	n.d.	n.d.	=	(Chen and Poon,
		superplasticizer					2017)
OPC	4.6	Sand	7	n.d.	n.d.	7	(Pan et al., 2003)
CEM I	4.6-6.9	Sand	7	7	n.d.	7	(Chang et al., 2010)
OPC	3.3	Sand	7	n.d.	n.d.	=	(Monzo et al., 1997)
OPC	4.7	Sand	n.d.	7	n.d.	7	(Mejdi et al., 2020)
CEM II/BL-	2.2 - 6.5	Sand	n.d.	n.d.	7	7	(Baeza-Brotons et
32,5R							al., 2014)
CEM II 42.5	2.2	Aggregate 0-4	7	7	n.d.	=	(Dražen Vouk et al.,
		mm					2017);



CEM III 32.5	4.4	Aggregate 0-4 mm + 5% metakaolin	7	7	n.d.	7	(Dražen Vouk et al., 2017);
CP-IV-RS-32	7	Quartz sand	7	n.d.	7	7	(Durante Ingunza et
CEM I 42.5 R	2.2	Quartz sand	n.d.	7	n.d.	7	al., 2018); (Piasta and
CEM I 42.5 R	5.1	Aggregate 0-2	n.d.	n.d.	n.d.	=	Lukawska, 2016) (Záleská et al., 2018)
CEM I 52,5	6.7	Silica sand	7	n.d.	n.d.	7	(Alcocel et al., 2006)
CEM I 52,5R	5.6 -11.1	Quartz sand 0–2 mm	7	7	n.d.	7	(Cyr et al., 2007)
CEM I 52,5N	2.2-6.7	Sand 0.516mm	7	n.d.	7	7	(Krejcirikova et al., 2019)
CEM II/A-LL 52.5R	4.4	River sand 2- 4mm	<i>\</i>	n.d.	n.d.	=	(Kappel et al., 2017).

- \* in a fresh state 264
- W workability 265
- S setting time 266
- 267 WA – water absorption of hardened samples
- CS compressive strength after 28-day of curing 268
- 269  $\searrow$  – deterioration of the parameter relative to the control sample
- $\nearrow$  improvement of the parameter relative to the control sample 270
- = no significant change in the parameter relative to the control sample 271
- 272 n.d. – no data

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Particles of SSA are characterized by considerable porosity causing the intensive water 274 absorption. The amount of SSA in the mortar mixture is a factor determining its workability 275 (Chang et al., 2010; Chen and Poon, 2017; S Naamane et al., 2016; Dražen Vouk et al., 2017). 276 The problem related to the deterioration of workability can be solved by milling, thanks to 277 which SSA particles become smoother, spherical-like and less porous (Chen et al., 2018; 278

Kappel et al., 2017; Monzo et al., 1997; Pan et al., 2003; Rutkowska et al., 2018). 279



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This phenomenon is called "lubricant effect" because the interlocking and friction between SSA particles are reduced (Pan et al., 2003). It is worth noting that the improvement of mortar workability by SSA milling is less significant compared to using a plasticizer (Monzo et al., 1997). Milling also can change the colour of the mortar to slight red if SSA contains significant amounts of Fe<sub>2</sub>O<sub>3</sub> (Kappel et al., 2017). Colour change, as well as significant shrinkage of the SSA-mortar during curing, may be limiting factors for its application (Li et al., 2017). The problem of workability can also be mitigated by water/binder (W/B) ratio reduction, but such solution can be used mainly for the production of prefabricates or soil stabilization approaches and not of joint mortar.

The presence of SSA in the mortar also affects its setting time. It was observed that the prolongation of the setting time is related to the amount of SSA in the mixture (Chang et al., 2010; Mejdi et al., 2020; S Naamane et al., 2016; Piasta and Lukawska, 2016). Extending the setting time (which is also longer compared to coal fly ash) is related to the presence of phosphates in SSA. Formation of insoluble calcium phosphate limits the access of water to the cement particles inhibiting the hydration reaction (Mejdi et al., 2020; Piasta and Lukawska, 2016). Sewage sludge ash milling which is performed to reduce excessive water absorption can also increase the setting time. The highly developed surface of SSA particles absorbs calcium ions thus the hydration process is inhibited (Pan et al., 2003).

The use of SSA in mortar mixtures may also cause corrosion of the reinforcement due to the high content of salts easily soluble in water. Alcocel et al. presented that the content of chlorides and sulfates in SSA was 7.8 mg/g SSA and 37.2 mg/g SSA respectively. Of course, the presented ratio will always strongly depend on the SS quality. However, it has been shown that the SSA rinsing and subsequent drying process limits its negative effect on the corrosion of reinforcement elements (Alcocel et al., 2006).

As mentioned, SSA obtained in the laboratory may show significant differences in both the oxide and mineralogical composition. The improvement of binding properties of mortars containing SSA with low CaO or amorphous phase content can be obtained by adding CaO to the mixture in the form of e.g. lime or pozzolanic cement. It was shown that mortar based on pozzolanic cement with the SSA addition (5.8% CaO in SSA mass) is water-tight, and thus capillary water absorption decreases with the share of SSA in the hardened mixture. The increase in mortar density, as well as the filling of free spaces between aggregate particles, improves mortar resistance to water and its mechanical properties. It was observed that samples containing 7% of SSA (in total fresh mortar mass) have 39% higher compressive strength in comparison to the control sample (Durante Ingunza et al., 2018).

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The mechanical properties of the mortars based on ordinary Portland cement even with a 5% addition of SSA with moderate CaO content (up to 20%) and a high proportion of the active amorphous phase did not differ significantly from the control samples. During mortar curing, the content of portlandite is reduced, but the pozzolanic reaction is not a factor conditioning the final properties of the material (Záleská et al., 2018). A slight decrease in the compressive strength of the mortar sample in relation to the control sample can be observed when SSA constitutes a greater proportion in the binder. Mejdi et al. reported that after 28 days of curing mortar with 20% cement replacement to SSA (containing 20,1% CaO) compressive strength of SSA-mortar was reduced by approximately 9% in comparison to control sample (Mejdi et al., 2020). Similar observations were made for mortar with high CaO content (31.3 - 52.2%), based on ordinary Portland cement. The presence of SSA in the mixture does not significantly reduce the compressive strength of mortars comparing to control samples. The use of SSA with high CaO content and maintaining an appropriate W / B ratio (0.5) by using plasticizer means that the mechanical properties of mortars are similar to reference samples, therefore there is no need for additional mineral materials or binders to improve mechanical properties (Monzo et al., 1997; Dražen Vouk et al., 2017).

Another solution to reduce the negative impact of SSA on the mechanical properties of mortars is the use of mineral additives (metakaolin, nano-silica) or multi-component cement. Nano-silica increases hydrates nucleation and crystal growth as well as accelerate the hydration reaction, which can improve the negative effects caused by SSA addition to mortars (Lin et al., 2008). Satisfactory strength properties are also obtained using multicomponent binders (e.g. CEM II, CEM III). These types of binders enhance the occurring pozzolan reaction thus obtained mortars are characterized by good mechanical properties (Baeza-Brotons et al., 2014; Kappel et al., 2017). Garcés et al. indicate that cement CEM II / B-M (V-LL) 42.5R is the most suitable binder for mortars containing SSA (Garcés et al., 2008), however, the use of additives or special cement must be preceded by an analysis of the SSA composition, because such solutions may not always be effective.

Replacing the main binder in mortars is usually limited to a small share of SSA. The production of mortars based on high-strength cement is an interesting solution. Despite the use of washed SSA (to remove corrosive ions), it was shown that with increased W / B ratio (0.7) and 60% cement (CEM I 52.5) replacement, a material with a compression strength of 13.0 MPa was obtained. Therefore the management of significant amounts of SSA is economically and environmentally justified and the resulting material could be used in construction (Alcocel et al., 2006). The use of SSA with a moderate CaO content, without prior

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removal of corrosive ions, as well as maintaining an appropriate W / B ratio results in a material with satisfactory mechanical properties. Despite the deterioration of strength in the early hardening phase, the mortar based on high-strength cement (CEM I 52.5) with 50 % binder substitution has a strength of only 16 % lower than the reference sample. However, it should be remembered that the final compressive strength of the mortar is not limited only to the use of high-class cement. Krejcirikova et al. presented research on the replacement of lime cement with SSA from two different SS incineration plants. Despite the use of SSA's with medium CaO content (23.4 and 36.4%) and high strength class cement, a significant decrease in strength was recorded with a 10% binder replacement (aprox. 25 and 20% respectively). It can be assumed that the SiO<sub>2</sub> / Al<sub>2</sub>O<sub>3</sub> ratio in both SSA's was high, therefore the deterioration of the compressive strength was mainly caused by the dilution effect and more porous structure formation (Krejcirikova et al., 2019).

As mentioned before, the hydrated structure of mortar or concrete can immobilize heavy metal cations (Záleská et al., 2018). It has been proven that the binding ability of Pb<sup>2+</sup> ions is connected with the content of SSA in the mortar. A high proportion of SSA in the mortar results in the reduction of the concentration of Pb<sup>2+</sup> ions in the leachate after washing the hardened samples (Li and Poon, 2017). The effect of immobilizing heavy metal ions in a hydrated cement matrix is related to the highly developed specific surface area, the porosity of the hardened material and the ion exchange effect induced by SSA particles. It has been observed that phosphates from SSA react with Pb2+ ions from the contaminated aggregate. The reaction product of both mentioned substrates is a crystalline lead phosphate (Li et al., 2017).

Most of the research on leaching heavy metals from hardened products are based on rinsing monolithic hardened mortar samples. In the case of crushed samples, it is obvious that the leachability of heavy metals is greater due to the larger contact surface. It has been reported that the increase in heavy metal concentration in crushed samples increased from 3 to 6 times compared to reference samples. However, the leaching behaviour of samples with SSA was of the same order of magnitude as the reference samples without residue (Cyr et al., 2007). It was shown that 25 % replacement of cement (CEM I 52,5 R) for SSA resulted in a 15 % reduction in the total mortar soluble fraction. The incorporation of SSA in mortar did increase the average concentration of heavy metals, but this difference in relation to the control sample is not statistically significant. Based on the analysis of the results, it was determined that the metal concentrations in the leachate: Ti, V, Cr, Ni, Cu, Zn, As, Cd, Sn, Sb and Pb meet the requirements for drinking water quality presented by the World Health Organization (WHO)

381 (Coutand et al., 2006). However, it is important to perform other elements leaching analysis to

prove full environmental safety of such an approach. 382

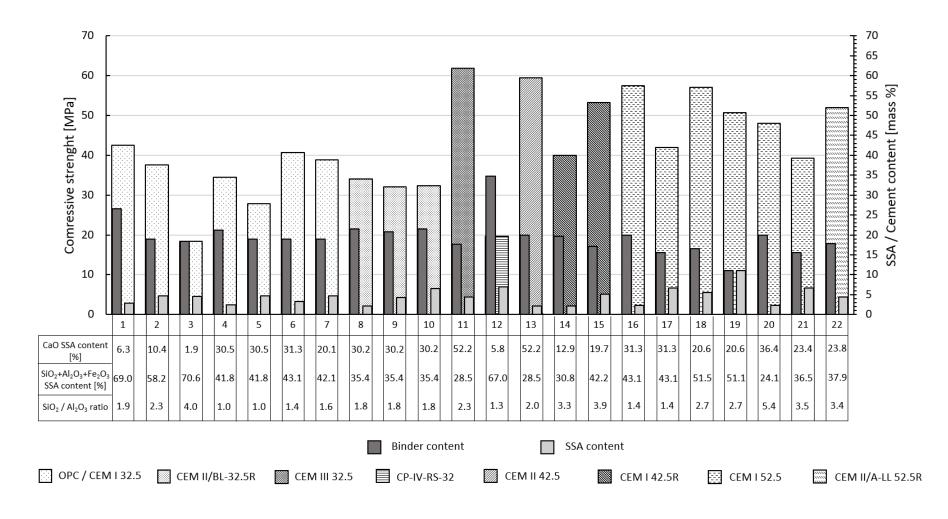


Figure 4. Compressive strength and shares of binders and SSA in mortars based on various cement binders after 28 days of curing. 1 - (Li et al., 2017); 2- (Chen and Poon, 2017); 3 - (Pan et al., 2003); 4, 5 - (Chang et al., 2010); 6 - (Monzo et al., 1997); 7 - (Mejdi et al., 2020); 8, 9, 10 - (Baeza-Brotons et al., 2014); 11, 13 - (Dražen Vouk et al., 2017); 12 - (Durante Ingunza et al., 2018); 14 - (Piasta and Lukawska, 2016); 15 - (Záleská et al., 2018); 16, 17 - (Alcocel et al., 2006); 18, 19 - (Cyr et al., 2007); 20, 21 - (Krejcirikova et al., 2019); 22 - (Kappel et al., 2017).

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Figure 4. shows the compressive strength of mortar samples in which SSA is present. Due to the diversity in the designed research, the Figure also shows the share of the binder as well as its type. To estimate the impact of SSA quality on the mechanical properties of mortars, the sum content of pozzolanic oxides  $(SiO_2 + Al_2O_3 + Fe_2O_3)$ , the  $SiO_2 / Al_2O_3$  ratio as well as the percentage content of CaO in SSA was also presented.

Based on the analysis of the data presented in Figure 4 it can be concluded that the quality of SSA has a significant impact on the mortars mechanical properties. When using Ordinary Portland Cement (OPC) and SSA with a low CaO content and high SiO<sub>2</sub> / Al<sub>2</sub>O<sub>3</sub> ratio (despite the high content of pozzolanic oxides), a material with low compressive strength was obtained (Chen and Poon, 2017; Li et al., 2017; Pan et al., 2003) despite the significant proportion of main binder (samples 1-3). In the case of using SSA with a moderate amount of CaO (approx. 20-30%) the mechanical properties of OPC-based mortars depend on the share of SSA and the SiO<sub>2</sub> / Al<sub>2</sub>O<sub>3</sub> ratio (samples 4-6) (Chang et al., 2010; Mejdi et al., 2020; Monzo et al., 1997). The deterioration of the mechanical properties of mortars may also be associated with the dilution effect, especially in the case of substitution of the main binder or SSA addition to the mixture without partial aggregate removing.

Pozzolanic cement as a source of CaO has a positive effect on the properties of mortars containing SSA. Durante Ingunza et al. presented that mortar compressive strength containing 7 % SSA was 24.7 % higher. Compared to the data presented in the Figure 4, mortar based on pozzolanic cement (sample 12) may seem unsuitable for this type of management methods, however, it obtains satisfying mechanical properties after longer curing time.

Mortars based on CEM II / BL-32.5R cement do not show significant loss of mechanical properties, even when significant amounts of SSA (over 5 %) are included in the mortar mass, which confirms the suitability of use multi-component cement as binders (samples 8-10) (Baeza-Brotons et al., 2014; Garcés et al., 2008).

As mentioned, the high content of CaO in the used SSA does not cause a significant loss of strength properties of the samples. The effect is more visible if the SiO<sub>2</sub> / Al<sub>2</sub>O<sub>3</sub> ratio is similar to the fly ash oxides ratio (approx. 2.4) (Fernández-Jiménez and Palomo, 2005; Jang and Lee, 2016; Richardson et al., 2016; Suksiripattanapong et al., 2015). Despite the low content of pozzolanic oxides in SSA and the different binders used (samples 11 and 13), satisfactory mechanical properties were obtained.

Based on the results presented by Piasta and Lukawska and Záleská et al. (samples 14 and 15) the addition of SSA with high SiO<sub>2</sub> / Al<sub>2</sub>O<sub>3</sub> ratio did not show a significant effect on the properties of the sample. The SSA used in the research can be treated more like a fine aggregate and not as an active mineral material (Piasta and Lukawska, 2016; Záleská et al., 2018).

In mortars based on high-strength cement (e.g. CEM I 52.5, CEM II 52.5), the SSA quality has the greatest impact on their properties, i.e. the amount of active oxides and their ratio (samples 16-22). The use of SSA with a moderate amount of CaO and ratio of oxides similar to coal fly ash allows to the incorporation of significant amounts of SSA (over 10 %) without significant loss of mechanical properties which in the context of waste management is the most appropriate solution.

# 4. Sewage sludge ash in concretes as a partial binder or fine aggregate substitute

Concrete is the second most commonly used material in construction after mortar. Research on the impact of SSA on concretes involves: main binder substitution, direct addition of SSA without removing the fine aggregate and partial replacement of the fine aggregate. The presence of SSA in concrete as well as in mortar reduces its durability (Chen et al., 2018; Fontes et al., 2016; Halliday et al., 2012). The SSA concrete strength loss is mainly proportional to SSA content (Jamshidi et al., 2013). Deterioration of mechanical properties is observed when SSA with a low CaO or amorphous phases content is introduced into the OPC concrete mixture (Fontes et al., 2016). It has been shown that substitution of 10 % of the main binder for SSA resulted in a slight deterioration of strength (4-8 %) compared to the control. Further increasing the SSA share (20 %) results in a 23-29 % strength reduction (Halliday et al., 2012). As presented by Lu et al. replacing the binder in 20% causes the binder dilution effect, therefore the compressive strength of the SSA concrete was lower than that of the control sample (Lu et al., 2019). It is worth adding that the introducing of significant amounts of SSA is associated with a significant loss of workability of the mix, therefore achieving homogeneity of concrete may become difficult (Halliday et al., 2012; Rutkowska et al., 2020). Changes in the basic parameters caused by the presence of SSA in the concrete mix are presented in Table 2.

Table 2. Characteristic of concrete samples containing SSA. 417

Type of	The share of	Type of aggregate	W	S	WA	CS	Reference
cement	SSA* [%]						
OPC	2.3-4.6	Granite <5mm	n.d.	n.d.	7	7	(Chen et al., 2018)
CEM I	1.5-4.5	Fine + 10 mm +	n.d.	n.d.	n.d.	7	(Halliday et al.,
		20 mm aggregate					2012)
OPC	1.8	Sand + gravel	n.d.	n.d.	n.d.	=	(Barbosa and Filho
							2004)
CEM I 32,5	0.7-2.2	Natural aggregate	n.d.	n.d.	n.d.	=	(Rutkowska et al.,
		0.125-16 mm					2018)
CEM I 32,5	0.8-3.9	Natural aggregate	n.d.	n.d.	n.d.	=	(Rutkowska et al.,
		0.125-16 mm					2020)
CEM II	1.6	Gravel and sand	n.d.	n.d.	=	7	(Jamshidi et al.,
							2012)
CP II F-32	1.2	River sand 2.8	n.d.	n.d.	7	7	(Fontes et al., 2016
		mm +					
		granite gravel 9.5					
		mm					
PC 32	2.9	Quartz sand +	7	n.d.	7	7	(Lima et al., 2015)
		granite gravel					
CEM I 42,5N	7.9-15.8	Sand 0/2, natural	n.d.	n.d.	=	7	(K.Kazberuk, 2011
- HSR/NA		aggregate 2/4 and					
		4/8					
CEM II BM	0.6-1.2	F-0/4 and F-2/8	n.d.	n.d.	7	7	(Baeza-Brotons et
(S-LL)-42.5R		aggregates					al., 2014).
CEM II BM	6.0	F-0/4 and F-2/8	n.d.	n.d.	7	7	(Baeza-Brotons et
(S-LL)-42.5R		aggregates					al., 2014).
CEM I 52,5	1,7	Natural aggregate	n.d.	n.d.	n.d.	7	(Lu et al., 2019)
		5-10 mm,					
		recycled					
		aggregates waste					
		glass aggregate 0-					
		5mm					



- 418 \* in a fresh state
- 419 W workability
- S setting time
- 421 WA water absorption of hardened samples
- 422 CS compressive strength after 28-day of curing
- $\searrow$  deterioration of the parameter relative to the control sample
- $\nearrow$  improvement of the parameter relative to the control sample
- = no significant change in the parameter relative to the control sample
- 426 n.d. no data

The presence of amorphous phase in SSA may enhance pozzolanic reaction in the later stages of concrete hardening, but mainly high CaO content and active oxide ratio in SSA intensify this reaction (Halliday et al., 2012). As the curing time increases, the compressive strength of the concrete sample increases, however, this increase is also observed for the control sample without the addition of SSA. By extending the curing time sufficiently long, the differences between the durability of SSA concrete and the control samples are insignificant (Rutkowska et al., 2020). This phenomenon can be related to the slight pozzolanic activity of SSA. Moreover, the SSA particles constituting additional nucleation sites during cement hydration (Jamshidi et al., 2012).

Sewage sludge ash obtained in an incineration plant with high CaO content can be successfully used as a partial binder substitute in concretes. It was shown that replacing 10 % of cement with SSA resulted in a 25 % improvement in compressive strength compared to control samples. It is possible that such products can be used in construction because their properties, e.g. strength, thermal conductivity or water resistance meet the requirements (e.g. EN 771-3 standard) (Pérez-Carrión et al., 2014).

The use of CaO-rich SSA, even if it is produced in a laboratory, as a partial replacement of fine aggregate increases the density of concrete, which results in increased strength. It has been observed that compressive strength of the concrete in which a part of the fine aggregate was replaced with SSA (30 % CaO) is twice higher than the samples in which the aggregate was not removed in favour of SSA (Baeza-Brotons et al., 2014). Rutkowska et al. reported that SSA containing a moderate amount of CaO (about 20%) can positively affect concrete properties. After 28 days of hardening of the concrete sample with the 20% SSA replacement, the compressive strength increased by 2.9% compared to the control (Rutkowska et al., 2020).

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The influence of SSA on the strength properties of concrete is probably caused by the pozzolanic reaction, since the substances contained in SSA and cement together may constitute a binder, which total amount is significantly higher. The problem associated with low CaO content in SSA can be solved by using pozzolanic cement to concrete preparation. It was found that the 3% SSA (6.2% CaO) addition (in total fresh concrete mass) without aggregate removal to concrete based on pozzolanic cement caused the hardened samples to be almost twice as durable as the control sample (Lima et al., 2015). Alternatively, CaO content can be increased directly in SSA, as mentioned before in section related to the SSA mortar quality.

The improvement of the mechanical properties of concretes is mostly observed in the case of a small proportion of SSA in the total mass of the cement mixture. Further increasing the content of SSA in the mixture results in high water demand during preparation and significant shrinkage of the products during curing (Chen and Poon, 2017; Rutkowska et al., 2018). Increasing the W / B ratio to improve the workability of the fresh concretes causes reduction in durability properties due to the formation of a more porous matrix. The use of plasticizer as well as milling or changing the W / B ratio is a good solution for the elimination of high SSA water absorption (Fontes et al., 2016; Halliday et al., 2012; Lima et al., 2015; Rutkowska et al., 2020, 2018). Ensuring homogeneity and water access to the binder particles does not significantly reduce the mechanical properties of SSA concrete (Jamshidi et al., 2013). It is worth adding that the SSA can increase concrete porosity, despite maintaining the appropriate W / B ratio and using a plasticizer. It has been shown that the porosity of concrete increases with the proportion of SSA in the mix, especially when low active SSA is used (Lu et al., 2019), however, the pores in the matrix are closed. The improvement of watertightness makes concrete more durable because it is more resistant to aggressive environments and frost (Baeza-Brotons et al., 2014; Barbosa and Filho, 2004). Moreover, SSA used as an admixture in concrete prefabricated/blocks, cause the thermal conductivity decrease due to reduced mix porosity and compactness (Lu et al., 2019).

The binding capacity of heavy metals by SSA is also confirmed with respect to the Commission Decision of 18 December 2014 (2014/955/EU) on the list of waste concerning, inter alia, the leaching of heavy metals from waste (Commission Decision of 18 December, 2014). For concrete in which cement was substituted with SSA in an amount of 15%, the sum content of elements in the leachate (Cd, Cr, Cu, Ni, Pb, Zn, As, Sb, Se, Ba, Hg, Mo) was 1.2 mg/L. A similar observation was recorded for 25% cement substitution, where the sum of mentioned leached heavy metals was 1.5 mg/L. In accordance with Commission Decision, the maximum concentration of each mentioned element in leachates cannot exceed 10 mg/l (Cieślik et al., 2018; Rutkowska et al., 2020, 2018). K.Kazberuk presented that replacing 25% of the binder in concrete for SSA, in addition to obtaining satisfactory strength properties, concrete can be regarded as safe and does not cause any environmental risk (K.Kazberuk, 2011). Based on heavy metal leaching tests (Toxicity Characteristic Leaching Procedure (TCLP Method 1311, 1992)) it was confirmed that the leachability of heavy metals from SSA-containing concrete was lower than in the case of coal fly ash-containing ones (Chen et al., 2018).

Figure 5. presents the compressive strength of concrete samples in which SSA is present as well as CaO content, the sum of active oxides and its ratio. Despite the high share of pozzolanic oxides and similar  $SiO_2$  /  $Al_2O_3$  ratio to coal fly ash, the mechanical properties of concretes (labelled as 1 and 2) are mostly determined by the share of binder and SSA, which in this case exhibits pozzolanic properties (Chen et al., 2018). Increasing active SSA content in concrete causes slight decrease in compressive strength, while lower  $SiO_2$  /  $Al_2O_3$  ratio in SSA causes more drastic strength loss with the increase of SSA's share (Halliday et al., 2012). The moderate content of CaO in SSA (about 20 %) may also contribute to the fact that the decrease in strength is less intense with the increasing share of SSA in concrete (samples 6, 7, 8).

Due to the limited data on the effect of SSA on CEM II-based concrete, its impact is difficult to determine. Considering the differences in SSA's composition (samples 9, 10), it can be assumed that the share of the main binder has the greatest impact on the concrete mechanical properties. Sewage sludge ash consisting mostly of SiO<sub>2</sub> has poor binding properties thus the decrease in strength is associated with a decrease in the overall binder content.

The results presented by Lima et al. where the concrete was based on pozzolanic cement, confirm that the external addition of CaO positively affects the SSA-concretes strength. It was shown that after 28 days of curing, in concrete with 10 % (in relation to the weight of cement) addition of SSA, the strength value improved by 44 % (Lima et al., 2015).

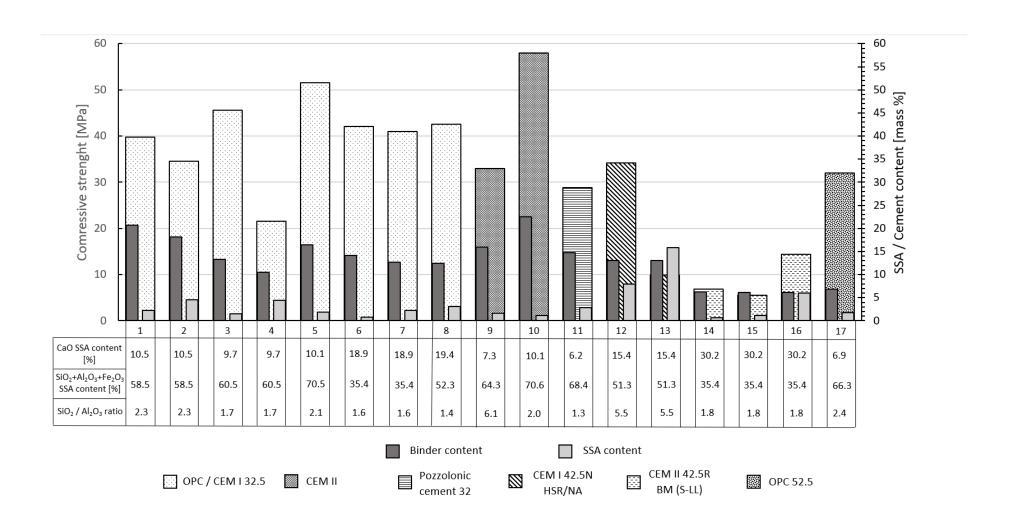


Figure 5. Compressive strength and shares of binders and SSA in concretes based on various cement binders after 28 days of curing.

1, 2 - (Chen et al., 2018); 3, 4 - (Halliday et al., 2012); 5 - (Barbosa and Filho, 2004); 6, 7 - (Rutkowska et al., 2018); 8 - (Rutkowska et al., 2020);

9 - (Jamshidi et al., 2012); 10 - (Fontes et al., 2016); 11 - (Lima et al., 2015); 12, 13 - (K.Kazberuk, 2011); 14, 15, 16 - (Baeza-Brotons et al., 2014); 17 - (Lu et al., 2019).

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The use of special binders (bridge and airport cement) in concrete with the addition of low-active SSA (samples 12 and 13) results in a drastic decrease in strength during the increasing share of SSA. It is worth noting that the values shown in Figure 5 relate to compressive strength after 28 days of curing. K.Kazberuk presented that after 180 days of curing, the samples are characterized by about twice as high strength as those after 28 days of curing (K.Kazberuk, 2011).

The test results labelled as 14-17 concern the use of SSA in concrete intended for the production of prefabricated elements. The nearly double increase in concrete strength may be associated with the pozzolanic reaction between SSA components and multi-component cement, causing secondary hydration phases formation, however, the increased density and filling the free spaces between aggregate particles by SSA was the decisive effect on final mechanical properties (Baeza-Brotons et al., 2014). Lu et al. indicate that such products do not require high durability, and therefore other waste materials can be included in the concrete mixture. In addition, production of partition blocks is a novel and eco-friendly approach for SSA utilization. (Lu et al., 2019).

It is worth mentioning that difficulty in estimating the impact of SSA on the physicochemical properties of hardened concretes is the variety of used aggregates. Stiffness and compressive strength of concrete depend also on the aggregate type as well as its particle size, therefore, differences between mechanical properties may occur (Ćosić et al., 2015).

5. Conclusions

Due to the increase in the popularity of SS thermal utilization methods, more and more ecological concepts of using SSA are being created. As a result of some similarities of SSA to coal fly ash, commonly used in the construction industry, SSA can be utilized in mortars or concretes mixtures. In such products, this waste is treated as an active pozzolanic additive or the main binder substitute. One of the greatest limitations of using SSA in cementitious building materials is the fact that SS from different incineration facilities may differ significantly. Regardless of the method of SS incineration, obtained SSA should always be treated as a material similar in properties to fine aggregate rather than a binder. Due to very diverse characteristics of SS, and its processing methods, SSA obtained from different sources should be thoroughly characterized separately before implementing the most appropriate utilization method. Moreover, the problem of the significant differences in the characteristics of SSA obtained at laboratory scale and SSA obtained at technical scale is crucial for developing of suitable waste management approach. Any strong statements regarding the possibilities of using

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SSA in the construction industry should be supported by at least SSA incineration industrialscale trials.

Based on the analysis of the results collected in this article, it could be stated that SSA, as a mineral component, has a negative impact on almost all mortar or concrete materials parameters. The activity of SSA in cement-based products depends mainly on its oxide and mineralogical composition as well as the type of used binder. The deterioration of mechanical properties is observed when low-active SSA even in small amounts is used. However, the negative impact of the presence of SSA on cement mixtures can be reduced by modifying the ratio of active oxides. The high share of CaO and pozzolanic oxides (SiO<sub>2</sub>, Fe<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>) in SSA may contribute to the pozzolanic reaction in a cementitious mixture based on OPC, thus compressive strength decrease is smaller. The discussed problem can be omitted at the stage of SS incineration. Addition of Ca, Si, Fe and Al species to SS is a trivial approach which could be an effective solution for the implementation of new waste management approach directly in the sewage sludge treatment plant or in the incineration facility. On the other hand, the use of low CaO content SSA could be more suitable when specific binders (e.g. multi-component cement) or additional source of CaO (lime or pozzolanic cement) are used. It is worth noting that the smallest parameters deterioration of SSA-containing samples is observed when highstrength binders (e.g. CEM I 52.5 or CEM II 52.5) are used. It is especially important in case of planning a possible increase of CaO content in SSA by simple technological operations at the stages of wastewater treatment or SS incineration.

The presence of SSA in cement mixtures, in addition to the deterioration of the mechanical characteristics, is also often associated with deterioration of workability, prolongation of setting time and product shrinkage. Worse workability parameter can be compensated by changing the W / B ratio, milling SSA or using a plasticizer. However, SSA milling can cause significant shrinkage of products during curing, while an increase in the W / B ratio is associated with an increase in porosity. The use of plasticizer is the most suitable, however, it can generate additional costs, especially when SSA constitutes a significant share in the product.

The last parameter determining both, durability and environmental safety of mortar and concrete is water absorption by cured samples. In the case of mortars, an increase in the water absorption capacity is observed while the concrete becomes more watertight. Therefore, the use of SSA in building materials should be limited mainly to applications in concrete mixtures.

It is also important to mention that the main advantage of incorporating SSA into building materials is the fact that the toxic heavy metals present in SSA are safely immobilized, and



the amount of unmanaged solid residues production for many sewage sludge treatment plants. It must be kept in mind that strength, workability, setting time or water absorption are not the only ones of the characteristic features that are affected by the presence of SSA in the cement mixture. For a detailed assessment, other aspects, such as environmental safety or other mechanical properties, should also be considered. Before considering any technological scale application, the laboratory-scale test should always be supported by data obtained during industrial-scale trials.

Despite the many negative aspects associated with the use of SSA in building materials, it is possible to reduce them by simple solutions but there is an urgent need for further research to optimize existing ones.

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# 6. Acknowledgements

This research did not receive any specific grants from funding agencies in the public, commercial, or not-for-profit sectors.

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