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Use of sludge generated at WWTP in the production of cement mortar and concrete

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Subject review

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Problems relating to disposal of sludge accumulating at waste-water treatment plants (WWTP) in Croatia are described in the paper. The possibility and adequacy of using ash obtained through incineration of sludge is considered. A special emphasis is placed on the significance of ash in concrete industry during production of cement mortar and concrete. Results of worldwide research conducted so far are described, with indications about the ways in which ash use influences properties of cement mortar and concrete.

Key words:
wastewater, WWTP, sludge disposal, ash, cement mortar, concrete

Pregledni rad

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Korištenje mulja s UPOV-a u proizvodnji cementnog morta i betona

U radu je opisana problematika vezana uz generiranje i zbrinjavanje mulja s UPOV-a (uređaja za pročišćavanje otpadnih voda) u Hrvatskoj. Poseban osvrt je dan na mogućnost i opravdanost korištenja pepela dobivenog spaljivanjem mulja. Naglasak je stavljen na ugradnju pepela u betonskoj industriji, pri proizvodnji cementnog morta i betona. U radu su opisani rezultati dosada provedenih istraživanja u okviru svjetske prakse, s primarnim osvrtom na utjecaj ugradnje pepela na karakteristike cementnog morta i betona.

Ključne riječi:
otpadne vode, UPOV, zbrinjavanje mulja, pepeo, cementni mort, beton

Übersichtsarbeit

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Anwendung von Schlamm aus Abwasserreinigungsanlagen in der Herstellung von Zementmörtel und Beton

In dieser Arbeit wird die Frage der Behandlung von Schlamm aus Abwasserreinigungsanlagen in Kroatien beschrieben. Die Möglichkeit und die Rechtfertigung der Verwendung von Asche, die bei der Verbrennung von Schlamm entsteht, werden betrachtet. Insbesondere wird die Bedeutung von Asche in der Betonindustrie bei der Herstellung von Zementmörtel und Beton erläutert. Die Resultate bisheriger weltweit durchgeführter Untersuchungen werden beschrieben und Auswirkungen des Einsatzes von Asche auf die Eigenschaften von Zementmörtel und Beton aufgezeigt.

Schlüsselwörter:
Abwasser, Abwasserreinigungsanlage, Schlammentsorgung, Asche, Zementmörtel, Beton

1. Introduction and legal framework

Proper water management is one of basic prerequisites for the development of any region. In addition, the need is now increasingly felt for proper protection of our environment, in the scope of which a special emphasis is placed on the protection of biodiversity and human health. This is why proper disposal of waste waters is ranked among high-priority activities in the scope of adequate water management policies. After analysis of the existing situation in the municipal services sector in Croatia [1-6], it can be concluded that, among all water management problems existing in Croatia, the greatest attention is now placed on adequate disposal of waste waters. This statement is further backed by the fact that only about 43 % of Croatian population is currently connected to a public sewerage system [6]. The situation is even less favourable with regard to connection to waste-water treatment plants (WWTP). According to available data [6], only 25 % of Croatian population is currently connected to plants ensuring second level of purification or higher. Here it should be noted that most of these plants have to be upgraded to the third level of purification as their capacity exceeds 10,000 equivalent population (EP), and they are situated in area defined as sensitive [7].

It is indicated in the Directive 91/271/EE, which came into force after Croatia's accession to the EU, that waste water must be collected and treated in all agglomerations greater than 2,000 EP. The second level of purification of all waste water must be provided for agglomerations greater than 2,000 EP, while higher levels of purification are required for agglomerations in excess of 10,000 EP in areas defined as sensitive and in their drainage basins (Council Directive 91/271/EEC concerning urban wastewater treatment). As in other EU countries this Directive with stricter quality requirements entered into force in 2005, Croatia now lags considerably behind most EU countries in the sphere of waste water purification practices. In an attempt to ensure compliance with provisions of the EU's Water Framework Directive, active measures are currently being taken relating to construction of comprehensive waste water drainage systems, with the corresponding waste water purification plants for agglomerations in excess of 10,000 EP. In other words, by joining the European Union, Croatia has assumed the obligation to build by 2018 all WWTPs with the capacity of more than 10,000 EP, which will finally result in the realization of the WWTPs for the total load of approximately 4,500,000 EP [8].

On the one side, construction of WWTPs exerts a favourable influence on the environment through reduction of waste substances that are released into environment but, on the other side, some new problems are encountered in the purification process, namely in the form of generation of significant quantities of sludge that must be adequately disposed. In other words, purification of waste water generates an additional type of waste called sludge (0.5 kg sludge/m3 of waste water; daily quantity of sludge dry matter varies from 50 – 70 g DM/EP·d). Adequate disposal of waste water (sanitary and industrial waste water in particular) implies collection, transport and purification of waste water but, at the same time, proper disposal of waste substances generated in the course of this purification process. According to Croatian legislation, the sludge generated at WWTPs is treated as non-hazardous waste that should be adequately treated and disposed into environment.

Current practices with regard to planning and construction of WWTPs are primarily oriented toward the water line, and attempts are made to meet the combined approach criterion according to which purified water must meet some specific purification requirements, and the discharge of purified water must not disturb natural biological balance in receiving water bodies (watercourses, channels, seas, etc.) [9]. The sludge disposal problem has not so far been fully solved in Croatia, nor has it been properly addressed through regulations, instructions or guidelines. As the construction of WWTPs gains in intensity in Croatia, the sludge disposal issue will increasingly become an encumbrance for public utility companies and local self-government units that operate local wastewater drainage and purification systems. The construction of WWTPs may be considered completed by the authorities only after a solution for final disposal of sludge has been provided, i.e. after compliance with all environmental protection measures has been demonstrated. This is directly related to obligations toward the EU because, in case all requirements – including adequate sludge disposal – are not met, significant funds will have to be paid in form of fines.

In 2007, the total annual generation of sludge originating from WWTPs amounted to almost 17,000,000 t DM in the USA and EU (namely 7,000,000 t in the USA and 10,000,000 t in the EU) [10]. Nevertheless, the quantities of sludge as a by-product of waste water purification are constantly on the rise all over the world. The annual sludge generation in 27 EU countries, including the sludge generation forecast until 2020, is presented in Figure 1 [11].

Almost 50 % of sludge generated in 2009 was used in agriculture in countries such as Spain, Great Britain and France, while the rest was either deposited on landfills or incinerated.

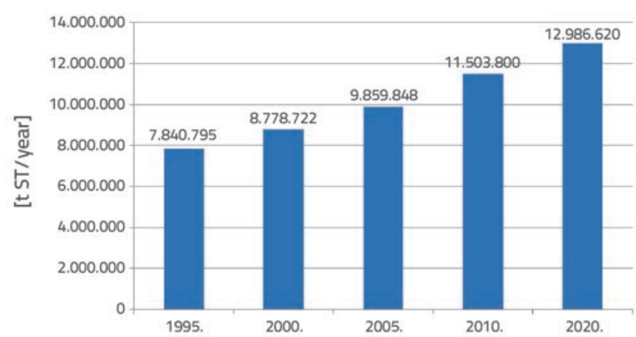

Figure 1. Annual sludge generation in WWTPs in 27 EU countries [11]

Sludge generated at the WWTPs that have been built so far in Croatia is deposited on solid waste disposal sites. Out of the total quantity of sludge, very little is used in agriculture and, for the

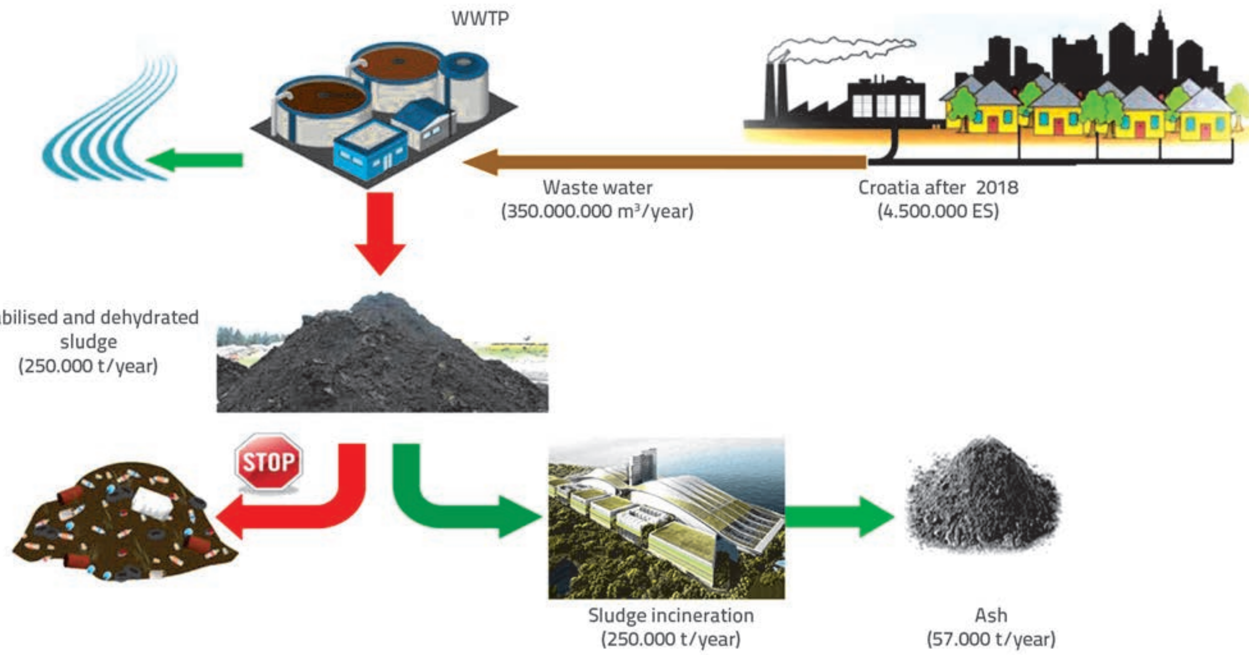

Figure 2. Estimated production of sludge and ash from WWTPs in Croatia after 2018

major part of this sludge, it can not be said with certainty where and how it is disposed. Until recently, the problem of adequate disposal of sludge has not been given the attention it deserves in Croatia. According to the *Byelaw on the ways and conditions for waste disposal, with categories and operating conditions for waste disposal sites* (OG Nos. 117/07, 111/11, 17/13), the disposal of sludge on waste disposal sites will no longer be allowed, starting from 2017. It is indicated in the same document that waste disposal sites will not be allowed to accept, inter alia, "municipal waste in case the mass of its biodegradable component exceeds 35 % of the total mass". The mass of the biologically stabilized sludge always exceeds 35 %wt of biodegradable matter. The criterion for the disposal of waste on non-hazardous waste disposal sites is also specified. This criterion defines limit values of no more than 5 %wt of dry matter for the total organic carbon (TOC) content, and no more than 5 %wt of TOC for stabilized sludge. It is obvious that the final sludge disposal problem will be gaining in significance as we are getting closer to the year 2017 and as more and more of new WWTPs are being built in Croatia. This will result in the need to finance and develop technologies through which sludge treatment and its final disposal will be solved in a different way [12], especially in the light of the fact that the startup of WWTPs amounting to 4,500,000 EP will result in generation of the total quantity of dehydrated and stabilised sludge of about 250,000 tons annually. As the quantities of sludge waste to be disposed on landfill sites is limited by European directives, namely by the EU Sewage Sludge Directive (86/278/EC), EU Landfill Directive (99/31/EC), and the EU Waste Incineration Directive (2000/76/EC), and as the EU Waste Framework Directive 2008/98/EC favours waste reuse (recycling) over other solutions, it is evident that the generation of such vast quantities of sludge originating from WWTPs will require an appropriate response.

Selection of an optimum sludge treatment procedure at WWTPs will inter alia depend on the final sludge disposal, and this final disposal must be taken into account already during construction of such plants. After a thorough analysis of various technical and technological solutions, and taking into account environmental protection costs, some studies [13, 14] conclude that the sludge incineration would be an acceptable concept of final sludge disposal in case of greater capacity WWTPs. Sludge incineration is compliant with the Appendix 1: Waste Disposal Procedures (D10) of the sustainable Waste Management Act (OG No. 94/13). The total sludge mass reduces considerably (up to 85 %) through incineration [15] and the same applies to the volume of the final waste matter, which facilitates further use of sludge and the newly created product (boiler/hearth ash). In addition, toxic organic compounds become disintegrated by thermal action, disagreeable odours are minimised, and the sludge energy can be put to good use [16]. However, a by-product in form of boiler ash and fly ash from filter is generated even during the sludge incineration procedure, and it also has to be disposed in an appropriate way. It should be noted that the quantities of such ash are far from negligible, with boiler ash quantities being about ten times greater compared to quantities of fly ash from filter. For instance, as much as 80,000 t/year of dehydrated and stabilised sludge will be produced at the central wastewater treatment plant in Zagreb, when the plant operation reaches full capacity (at 1,500,000 EP), while about 18,000 t/year of ash would be generated by sludge incineration, out of which about 16,500 t/year of boiler ash and about 1,500 t/year of fly ash from filter [17]. A schematic of sludge production in Croatia after 2018 is presented in Figure 2. An estimate of ash production is given under assumption that all sludge generated in the process will be incinerated.

Donatello and Cheeseman [18] estimate that approximately 1,700,000 tons of ash are generated annually on the global level during thermal treatment of sludge from WWTPs (mostly in the US, EU, and Japan). This number is expected to rise steadily due to construction of new and renovation of extisting WWTPs. Research conducted so far has shown that, due to its characteristics and chemical composition, the ash created in this way can be used in some branches of economy, especially in construction industry where it can be used in the production of cement, concrete, bricks, ceramics, asphalt mixes in road construction, soil improvement mixes, and for extraction of phosphorus, which is a limited resource on the Earth [10, 15, 18-27].

From the perspective of sustainable development, the waste water treatment cycle is almost fully completed by the use of sludge, and the quantities of waste matter generated in the process, which still have to be disposed into environment, are quite negligible.

In this paper, a primary focus will be placed on the use of ash in concrete industry, as concrete is the world's most widely used artificially obtained material. Some practical results of research conducted in various countries will be described, and comparison will be made with the results obtained in Croatia.

2. Composition of ash obtained by incineration of sludge

In concrete industry, the use of ash obtained by incineration of sludge is primarily related to the ash from boilers. Ash is a powdery material with some sand size grains, and with a negligible content of organic matter and moisture. An accurate range of ash grain sizes depends on the sludge treatment procedure, on the content of industrial water in waste water, and on the type of drainage system [18]. The ash grain size can vary from 1 to 100 mm, with an average size of approximately 26 mm [28, 29], and in some kinds of ash (even up to 90 %) it can be less than 75 mm [29]. The irregular morphology and porous structure of ash grains, and its large specific surface, result in a greater need for water when the ash is used as a replacement in various types of cement mortars and concretes [10].

Principal chemical elements contained in ash obtained during thermal treatment of sludge are the silicon (Si), aluminium (Al), calcium (Ca), and phosphorus (P). According to some authors, the ash density varies (but not to a great extent) from 2.3 to 3.2 g/cm^3 [27], 2.62 g/cm^3 [30], and 2.86 g/cm^3 [31]. Densities of ash obtained by incinerating sludge from Karlovac WWTP at various temperatures are presented in Table 1 [8].

Table 1. Density of ash obtained by incineration of sludge from Karlovac WWTP at various temperatures (800, 900, and 1000 °C) [8]

Incineration temperature [°C]	Density of ash [g/cm^3]
800	2,67
900	2,73
1000	2,83

Heavy metals such as mercury (Hg), cadmium (Cd), arsenic (As) and lead (Pb) should be burnt by incineration or separated from the ash with exhaust gases, and retained on the filter with fly ash [32]. However, traces of metal may remain in ash due to their condensation on ash particles during decrease of temperature in the incinerator.

It should be noted that there are many positive experiences in the world relating to the use of ash obtained during the sludge incineration process. However, some research projects are still in progress, and new projects of this type

Table 2. Chemical composition of ash obtained by incineration of sludge [8, 10, 16, 23, 28, 31, 34-44]

Chemical composition	Oxide content in ash [mas. %]			
	Range of values (literature review)	WWTP Karlovac	WWTP Koprivnica	WWTP Zagreb
Fe_2O_3	4,70 - 20,00	8,21 - 9,46	0,31 - 0,40	4,32 - 5,99
SiO_2	17,27 - 50,60	2,87 - 7,94	0,41 - 0,49	16,21 - 22,12
CaO	1,93 - 31,30	37,64 - 42,12	92,82 - 93,83	39,18 - 52,23
MgO	1,40 - 3,22	4,23 - 4,53	0,71 - 0,77	2,98 - 3,45
Al_2O_3	6,32 - 19,09	11,72 - 16,46	0,89 - 1,19	7,97 - 10,83
P_2O_5	1,67 - 18,17	16,02 - 17,21	0,80 - 0,83	5,21 - 7,54
TiO_2	0,29 - 1,00	0,76 - 1,03	0,04 - 0,05	0,75 - 0,97
Na_2O	0,32 - 1,26	0,28	0,03 - 0,04	0,12 - 0,22
K_2O	0,62 - 2,34	1,26 - 1,31	0,12 - 0,13	0,36 - 0,78

are constantly being initiated. Sludge (ash) use possibilities are greatly dependent on its chemical composition. Lopes and al. [33] and Chen and al. [27] have determined that the composition and properties of ash are greatly influenced by the origin of wastewater, by the type and quantity of additives during their treatment, and by the sludge treatment method. This points to the great significance of the ash quality and composition testing under various circumstances, as related to the quality of wastewater and technological processes used at the WWTP. That is why the results of some research conducted with the sludge whose composition differs from that of the sludge generated in the territory of Croatia should be taken with some caution. For instance, industrial production decreased considerably over the last two decades in Croatia, which resulted in a considerable change in the quantity and composition of wastewater treated at WWTPs, and hence also in the composition of sludge. Similarly, the composition of sludge and ash generated at the WWTP depends to a great extent on the technological process used in water purification and in subsequent treatment of sludge. To provide a better insight into differences in ash composition and properties, some percentage values (%wt) are presented in Table 2 for individual chemical compounds (oxides) in ash obtained by incineration of sludge at various WWTPs in the world [10, 16, 23, 28, 31, 34-44]. To enable comparison, the same table provides weight percent values of the same oxides in the ash obtained by incinerating sludge from three WWTP in Croatia (Karlovac, Koprivnica and Zagreb [8].

Table 2 shows that there are considerable differences in ash composition. These differences primarily depend of the properties of wastewater, on the sludge treatment technology, incineration temperatures, etc. For instance, the patented "MID-MIX" sludge treatment technology is used at the Koprivnica WWTP. This technology is based on addition of considerable quantities of lime during sludge treatment, and so the ash obtained from this sludge has a much higher CaO content. Ranges of values for oxides contained in ash from the three WWTPs in Croatia are due to different sludge incineration temperatures (800, 900 and 1000°C). It has however been observed that the proportion of principal oxides contained in ash may vary considerably even at plants operating under similar conditions [45, 46]. The content of elements that are present in ash in traces (or in smaller proportions) may vary to an even greater extent because of the nature of industrial activity in a particular area.

3. Incineration of sludge

The drained sludge, with the proportion of dry mater varying from 18 to 35 %, is normally dried prior to incineration so as to increase the incineration efficiency. The sludge dried in this way, with the dry matter content of 75-90 by weight, is then introduced into incinerators where the ash in form of a finely granulated waste material is created. The incineration of sludge originating from WWTPs does not only result in creation of ash that can be used in a number of ways but. It also results in a thorough thermal disintegration of organic and most inorganic pollutants [47].

Although the calorific value of sludge is similar to that of the brown coal, it should be noted that this is the calorific value of the organic portion of sludge, while the inorganic part has no calorific value. That is why, prior to incineration, the sludge has to be treated (drained) to the dry matter content varying from 28 to 33 %wt in order to enable spontaneous combustion of sludge without adding external fuel to maintain the combustion process [18]. In this stabilized and dehydrated state, the calorific value of sludge ranges from 12 to 20 MJ/kg [36].

Several incinerator types are currently used for the incineration of sludge. Most widely used systems are the fluidized bed incinerators [10, 15, 18, 30, 39] whose schematic is given in Figure 3. Other types currently in use are modular furnaces [40] and electrical muffle furnaces [16].

As shown in Figure 3, the sludge and hot air (500 - 600°C) are introduced into the furnace. The temperature in the furnace is controlled by "injecting" water or liquefied petroleum gas. The role of sand placed at the bottom of the furnace is to assist in stabilisation of temperature changes within the furnace itself.

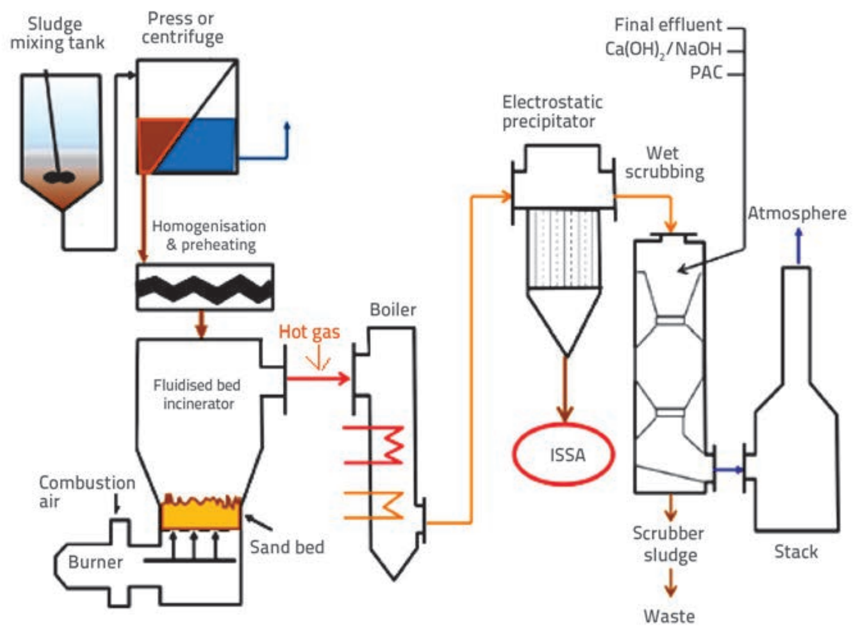

Figure 3. Fluidized bed incinerator [18], where: ISSA is the incinerated sewage sludge as and PAC is the polyaluminium chloride

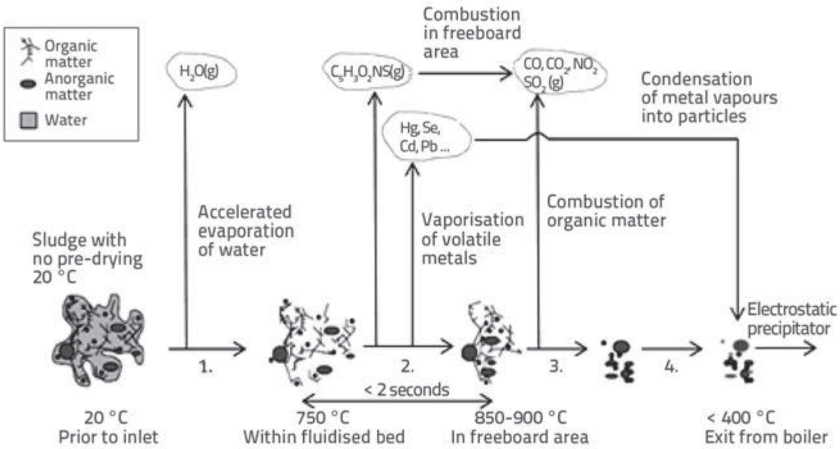

Figure 4. Physical and chemical processes during incineration [18]

The sludge is incinerated in the furnace for 1 to 2 hours. During this time, the water and easily volatile metals are evaporated, and organic components are fully combusted, resulting in the formation of gases. The remaining inorganic substances are extracted from the furnace, together with flue gases, in form of fine particles. The ash formed in this way (ISSA) passes through the heat exchanger (boiler), and is retained in bag filters or electrostatic filtering devices. Before being discharged into the atmosphere, flue gases are treated in accordance with the EU directive on incineration of waste (2000/76/EU [18]. Physical and chemical processes taking place during incineration of sludge are presented in Figure 4.

The ash properties, such as density and water absorption, are greatly influenced by the sludge incineration temperature, which may consequently influence the porosity, mineralogy, and microstructure of the final product in which the ash is incorporated. A variety of influences of the sludge incineration temperature on the produced ash properties has been observed by various authors. The increase of incineration temperature from 800 to 900°C results in a reduced water absorption (water absorptions decreases significantly above 1000°C) and in the ash weight reduction (due to additional oxidation of organic matter), which in turn results in higher density (specific mass) of the ash, the maximum values being registered at the temperature of 1000°C [48]. Depending on the sources of information, clinker is created at temperatures in excess of 900 - 950°C, i.e. when an increase in crystallisation and weakening of pozzolanic properties are registered [15, 16, 31].

The temperature range at which influences of the sludge incineration temperature on ash properties are analysed varies from 450 to 1300˚C. Tantawy et al. [16] have concluded that, considering the need to preserve pozzolanic properties of ash, an optimum sludge incineration temperature amounts to approximately 800 ˚C. During incineration, it is sometimes possible to add certain chemicals, such as lime [15], so as to improve properties of the resulting ash.

4. Adding ash to cement mortar and concrete

4.1. General

Concrete is currently the world's most widely used artificial building material. Mineral concrete additives are defined as inorganic materials, pozzolanic materials and/or materials with latent hydraulic properties which, when finely ground, can be added to concrete and/or cement mortar so as to improve some specific properties or to achieve some targeted properties [49].

According to published research results [18, 29], there is a great potential in the concrete product industry for replacement of some concrete components (cement, aggregate) with appropriate materials, including the ash resulting from thermal treatment of sludge originating from WWTPs. However, conditions, methods and quantities that can be replaced depend on a number of factors, and must be analysed individually for each type of replacement component [27]. For instance, the influence of sludge incineration conditions (type of furnace, incineration temperature, water content in sludge (by mass), incineration time) on the reduction of heavy metal content in ash is analysed so as to develop a more efficient and environmentally acceptable sludge incineration procedure.

Considering its composition and pozzolanic activity, the greatest potential for the use of ISSA in the production of cement mortar and concrete, lies in the replacement of a certain weight proportion of cement. Many research results have been published over the past decade in this field. Some of them are presented in the following sections of this paper. The second less studied way of using ash in concrete industry involves a partial replacement of fine aggregate [50]. Concretes in which up to 30 %wt of sand is replaced with ash exhibit the compressive strength reduction of 22 % in the hardened state after 28 days [51]. A higher demand for water due to a more porous structure of ash, compared to sand, limits the proportion of sand replacement with ash to <5 - 10 %wt [18]. After analysis of research activity in this area, it can be concluded that possibilities for using ash as replacement for fine aggregate have not been studied in great detail.

4.2. Pozzolanic activity

Many studies have revealed that the ash obtained by incineration of sludge (ISSA) exhibits a certain level of pozzolanic activity. According to their definition, pozzolans are materials that contain SiO_2 or SiO_2 and Al_2O_3. Taken by themselves, pozzolans do not have any binding capacity, but in a finely dispersed form and in the presence of water they react chemically with lime at room temperature and form compounds that present binding properties. A considerable SiO_2 and Al_2O_3 content, and a great specific surface (Tables 2 and 3), indicate that the ISSA could be used as a pozzolanic material. The pozzolanic activity of ash is usually evaluated by direct methods (which register reduction in $Ca(OH)_2$ content in the lime-ash mix during pozzolanic reaction) and indirect methods (which measure changes in physical properties, mostly compressive strength of standard cement mortars with replacement ash during hydration reaction) [10]. So far, most conclusions about the reactivity of ash have been made using indirect methods, through determination of compressive strength. The observed negative influence of ash on strength development, in cases when ash is used as a partial replacement of cement, is mainly attributed to an increased need for water due to irregular morphology of ash particles [18]. Using the direct method involving application of thermogravimetric analysis (TGA), Jamshidi at al. [52] have determined the pozzolanic activity of ash in the amount of 37.86 %, which is much lower compared to the activity of natural pozzolans (70 %). According to the research made by Fontesa et al. [37], the ash with the total content of basic oxides (SiO_2+Al_2O_3+Fe_2O_3) of more than 70 % by weight can be used as pozzolanic material in the mix containing Portland cement.

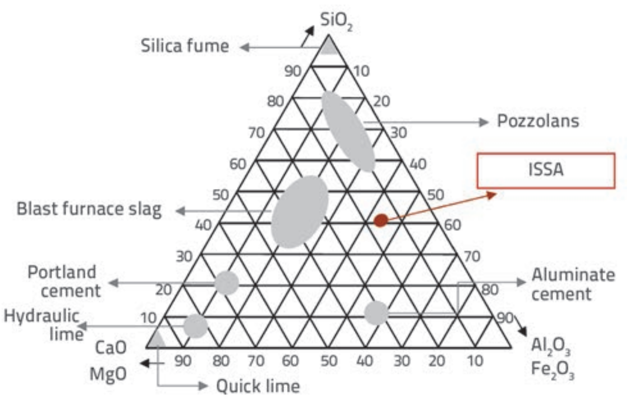

Figure 5. Ternary Keil-Rankin diagram for the CaO-SiO2-Al2O3 system with the position of various mineral additives to cement and cement types, where ISSA is the incinerated sewage sludge ash [27]

The ternary Keil-Rankin diagram for the CaO-SiO_2-Al_2O_3 system, with the position of ash (ISSA) as related to the positions of various mineral cement additives and cement types, is presented in Figure 5. In the presence of pozzolanic materials, cement exhibits much greater pozzolanic activity compared to other binding materials because, during hydration, it liberates the calcium hydroxide ($Ca(OH)_2$) that is needed for pozzolanic reaction (reaction between ($Ca(OH)_2$ and SiO_2 from pozzolan). Today we have many methods for activating low-reaction binders (annealed clay, fly ash from coal-powered thermal power stations, granulated high-furnace slag). The most common one is the method involving chemical activation by alkalis (NaOH, KOH or Na_2O_3Si), during which the so called geopolymers (alkali-activated aluminosilicates) are created. As some chemical activators are highly corrosive and may affect the safety of workers, the current trend is to use less aggressive substances or to add smaller quantities of cement as activator. The future research in this area should be oriented toward finding an optimum activation method for the ash generated by sludge incineration. The reactivity of the locally available sludge-based ash should be considered from the aspect of its microstructure, chemical and mineralogical composition, and pozzolanic activity, and this at various thermal treatment conditions, adding various additives during the thermal treatment process and during concrete mixing.

4.3. Influence of ash on fresh concrete properties

A partial replacement of Portland cement with ash affects both the concrete workability and strength development [10, 18, 30, 39, 40, 52]. In cases when parts of basic concrete components were replaced with ash originating from thermal treatment of sludge, a longer setting time and lower strength of concrete components obtained in this way was registered [18, 53]. According to Gunnu et al. [54], the replacement of 20 % by weight of cement with ash causes delay in the start and end of setting time, so that this time is extended by 2.5 times as related to the reference mix. This can among other things be attributed to additional quantity of water in mixes with a greater ash content, where the water had to be added to maintain workability. The results of first studies involving ash obtained from sludge from the Karlovac WWTP reveal delay in the start and end of mortar setting, with a higher sludge incineration temperature. Here, the longest setting times were obtained for mixes with 10 and 20 % by weight of ash (total analysed ash proportions ranged from 0 to 30 % by weight) [8]. Irregular morphology of ash particles results in a lower workability of concrete even at small proportions of concrete replacement with ash, which is primarily due to an increased need for water. When testing influence of ash addition (0-30 %wt) on the workability of cement mortars, Monzo et al. [39] observed that the workability decreases nonlinearly with an increase in the proportion of ash, and so the decrease in workability is less pronounced when a great proportion of ash is used. Studies involving ash obtained at the Karlovac WWTP [8] show that the workability of cement mortars containing ash improves with an increase in sludge incineration temperature (Figure 6).

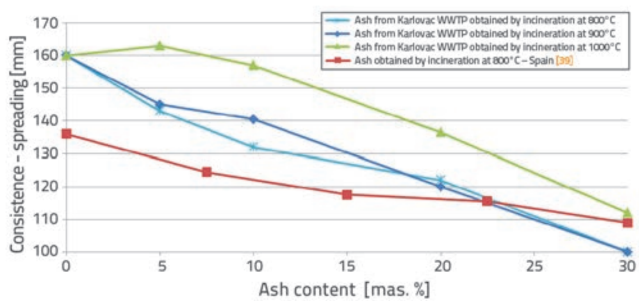

Figure 6. Workability of cement mortar as related to ash content and incineration temperature: comparison of results obtained at Karlovac WWTP [8] with previous studies [39] (w/c = 0,50; natural 0-4 mm aggregate)

Table 3. Influence of grinding time on fineness, density and specific surface of ash [40]

Property	Ash grinding time [min]						
	10	20	30	60	120	180	360
Blaine fineness [m²/kg]	496	780	846	975	979	993	872
Density [g/cm³]	2,48	2,61	-	2,54	2,45	2,67	2,60
BET specific surface [m²/kg]	11,588	10,906	11,774	11,020	11,666	11,521	12,487

According to some authors, poor workability can be improved by increasing ash fineness [40], and this either by adding a superplasticiser, Figure 7, [39] and/or by adding fly ash from coal-powered thermal power plants [41]. Pan et al. [40] concluded that an increase in the ash grinding time (from 10 to 360 min) influences the increase in the ash fineness and its pozzolanic activity, but that it does not bear a significant influence on the change in density and specific surface of the ash (Table 3). An increase in ash fineness also results in an increase in the cement mortar strength, improvement of workability, extension of setting time, and better water absorption (due to greater free surface area of ash particles). Nevertheless, according to the study of total porosity of cement mortars and concretes prepared with replacement ash (5 to 30 %wt) conducted by Fontes et al. [37], a significant increase in the total porosity was observed in mixes in which the ash proportion amounted to 10 % by weight or more.

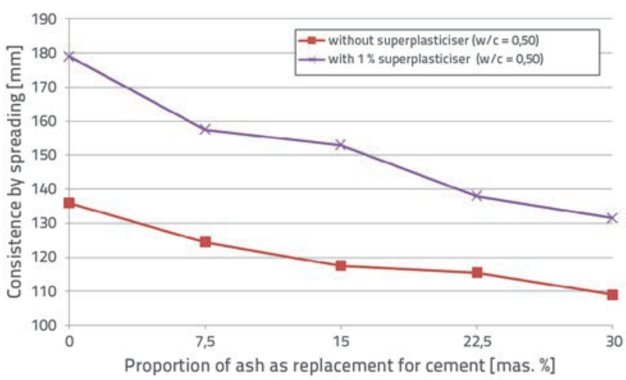

Figure 7. Workability of cement mortar with and without 1 % of superplasticiser as related to the content of replacement ((0 – 30 %wt) [39]

A significant need for water, resulting from reduced workability and extension in setting time in fresh cement mixes prepared with ash, may exert a significant influence on the dimensional stability of ash-containing concrete. Therefore, further research should focus on testing deformations that occur during first hours of hydration, and on long-lasting deformations occurring due to concrete drying. In addition, further testing of combined action of various ash-containing binding materials is also needed, with regard to acceleration of setting time, and hence a faster increase in strength.

4.4. Influence of ash on hardened concrete properties

The research carried out so far on the influence of ash on mechanical properties of cement, mortar, and concrete has confirmed that the compressive strength reduces with an increase in the ash content by weight, while an increase in ash fineness contributes to the concrete strength when similar quantity of replacement ash is used. However, when results obtained by individual authors are compared, significant differences can be noted in relative values of the results (fall in ash-containing sample strength compared to control samples). For instance, if 20 %wt of Portland cement is replaced with ash, the compressive strength can be reduced by 32 % [55], 52 % [40], and 24 % [56]. Donatello and Cheeseman [18] consider that these differences in results are due to the difference in cement properties, W/c ratios, sample dimensions, etc., but that they are also the result of the difference in the processes used during treatment and incineration of sludge (incineration temperature for instance) and the difference in the resulting ash composition, which is the area in which most future research should be concentrated. Cyr et al. [10] have observed that the negative influence of ash on the development of early strength of concrete (up to 7 days) reduces considerable in subsequent hardening phases (after 28 days).
Jamshidi et al. [52] have observed that the compressive strength of concrete reduces by 8 % if 10 %wt of replacement ash is added. Based on these authors, more than 10 %wt of replacement ash should be used only in case of non-structural concretes. According to Chen et al. [27], the compressive and bending strength of analysed cement mortar and concrete samples containing replacement ash (10 – 30 %wt) decreases linearly with an increase in the replacement ash content in cement mixture. The authors explain this with two hypotheses: a greater need for water in cement mixes containing replacement ash (in order to ensure proper workability), and poor hydraulic properties of ash (compared to cement) due to lower CaO content in ash (less than 10 %wt). According to the same authors, an optimum replacement of basic concrete constitutes with ash is: 10 %wt of ash as replacement for cement and 2 %wt of ash as replacement for sand. Fintes et al. [37] tested the influence of replacement ash (10-30 %wt) on the development of compressive strength of cement mortars and high-performance concretes, and concluded that the concrete

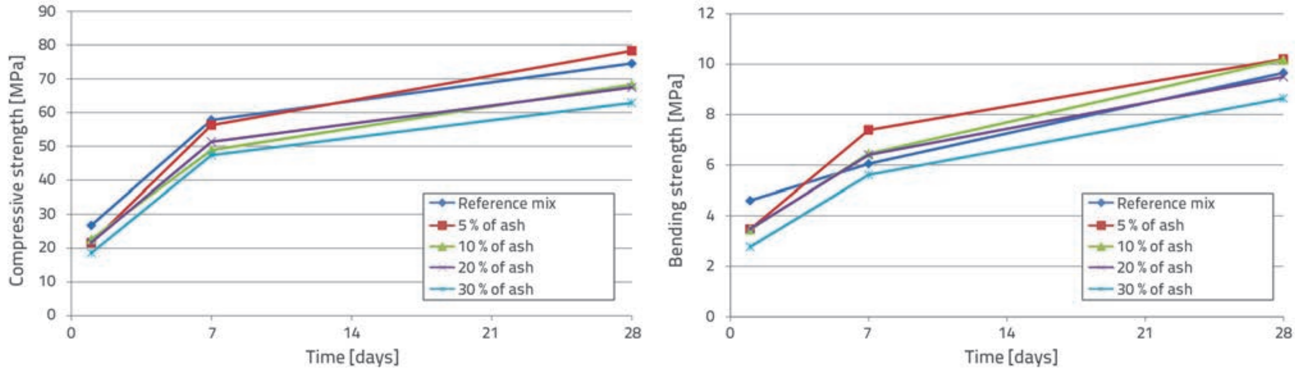

Figure 8. Compressive and bending strength of cement mortar as related to the content of replacement ash (0 – 30 %wt) obtained by incineration of sludge from the Karlovac WWTP (w/c=0.50; sludge incineration temperature 900°C) [8]

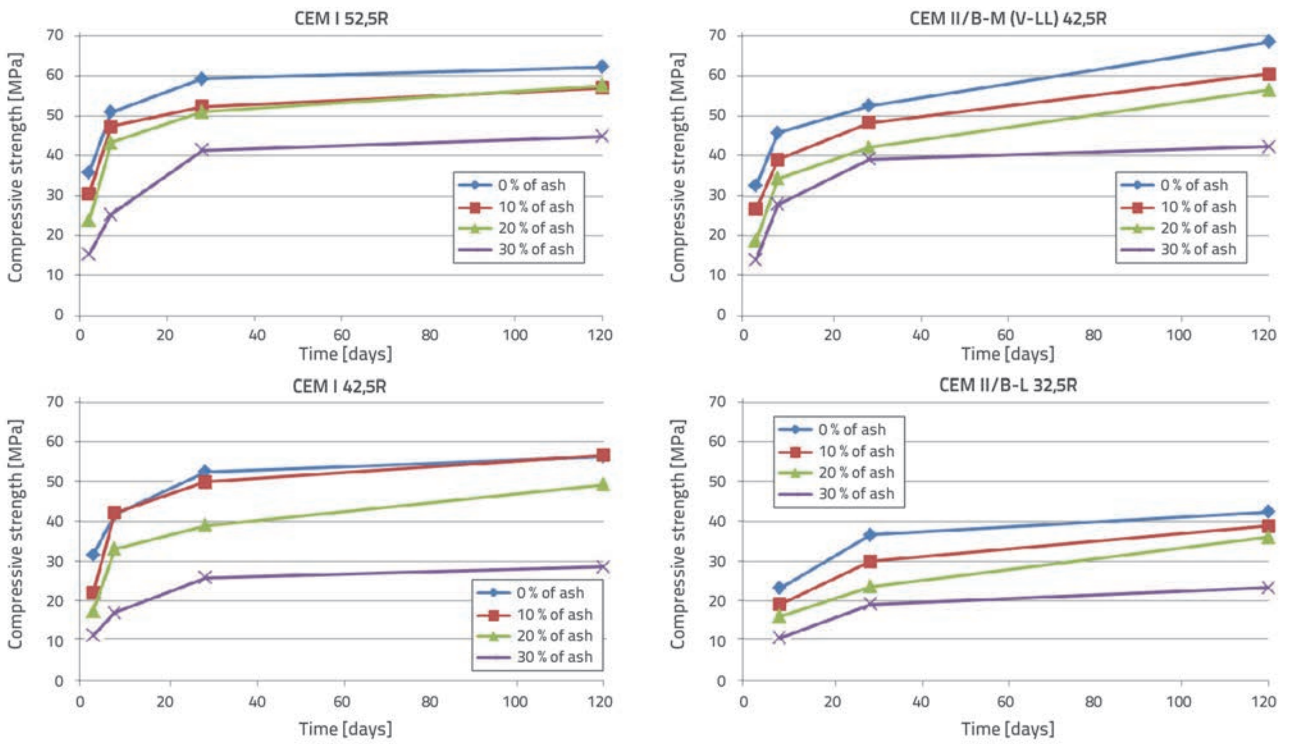

Figure 9. Compressive strength of mortars as related to the type of cement and %wt of ash [30] (where CEM I 42,5/52,5R is the ordinary high early strength Portland cement, CEM II/B-M (V-LL) 42,5 R is the high early strength Portland cement with silicate fly ash and limestone, and CEM II/B-L 32,5N is the normal early strength Portland cement with limestone)

strength obtained with 5 to 10 %wt of ash are satisfactory for the control sample class. When studying influence of replacement ash (15 and 30 %wt) on the development of compressive strength of cement mortar samples (cured in water at 40°C) during 28 days of hydration, Monzo et al. [57-59] observed a moderate increase in compressive strength of the analysed samples (due to pozzolanic properties of ash) as compared to control samples [59]. In addition, these authors registered a 15.3 % increase in compressive strength after 28 days of hydration for cement mortar samples with 15 %wt of ash and with a superplasticiser [57]. First studies conducted in Croatia involved cement mortar with the replacement of cement by 5, 10, 15, 20 and 30 %wt of ash obtained by sludge incineration (800°C, 900°C, and 1000°C) at the WWTP in Karlovac. The cement CEM II/B-M(S-V) 42.5N, and the dolomitic sand (grading: 0.4 mm) were used for preparation of a series of mortar samples (w/c = 0.45, 0.50, and 0.55) [8]. According to this research, depending on w/c ratio of cement mortar samples and sludge incineration temperature, the mixes with 20 %wt of ash (w/c = 0.50; incineration temperature = 900°C) exhibited even grater compressive and bending strength compared to the reference sample (mortar without ash) for all hydration times considered (1, 7, and 28 days). Some results obtained during this research are shown in Figure 8. The analysis of bending and compressive strength results shows that the strength of cement mortar samples with 5, 10, 20, and 30 %wt of ash (w/c

= 0.50; sludge incineration temperature: 900 °C) increases with hydration time, and decreases with an increase in replacement ash content. Mortar samples with up to 20 %wt of ash exhibit greater or equal bending strength compared to the reference mortar sample already after seven days of hydration. After 28 days of hydration, the mortar sample with 5 %wt of ash exhibits the greatest compressive strength, even exceeding that of the reference sample.

Some international research results have revealed great differences in the influence of ash on mechanical properties of cement mortar or concrete. These differences are primarily due to the difference in size of samples and to different w/c ratios used on individual research projects [18]. In addition, a significant influence is also exerted by the process used for thermal treatment of sludge: type of furnace, temperature, influence of various additives during thermal treatment, etc. A review of the current state-of-the-art in this field shows that the influence of thermal treatment on physical and chemical properties of ash, and the concrete/concrete elements preparation and treatment technology, have not so far been fully investigated, and that an ample space still exists in this area for further research. It would be especially worthwhile to study the possibility of combining binding materials as these materials could, together with ash obtained form sludge, develop better mechanical properties compared to the current research results. Garces et al. [30] have shown that the type of commercially available cement (ordinary or mixed cement) also influences mechanical properties of cement mortars in which 10 to 30 %wt of cement is replaced with ash. According to this research (Figure 9), compressive strength of all mortar samples increases with hydration time, and decreases with an increase in the replacement ash content. Strength values developed in this way are, generally, lower compared to the corresponding reference samples (not containing ash). Mortar samples with 10 %wt of ash exhibit the greatest compressive strength values, regardless of cement type. According to these authors, when compared to cement types used, the mixed cement CEM II/B-M (V-LL) is the most appropriate for the preparation of ash-containing mortars. After 120 days of hydration, these samples have the same or greater compressive strength values compared to mortar samples prepared with the CEM I 52.5R cement (cement exhibiting the greatest nominal strength). The authors state that this is due to the synergic effect of fly ash (cement ingredient) and ash added to the cement mix.

4.5. Long-term behaviour

The environmental impact of adding ash to concrete has been studied by several authors. The so called leaching test has been used to determine concentration of heavy metals leached from concrete elements to which ash is added. The results obtained confirm that the concentration of potential pollutants (heavy metals in particular) present in ash is within allowable limits [18, 27].

It has been established by review of situation in this area that most authors focus their research exclusively on changes in mechanical properties of concrete, and partly on the leaching of heavy metals. Considering the growing significance accorded to durability of concrete elements, further research should also concentrate on the influence of replacement ash, i.e. on the combined influence of ash and other binding materials on the long-term behaviour of concrete elements in aggressive environments, which is especially important for the elements that are continuously or occasionally exposed to water action.

5. Conclusions

The problem of sludge disposal is currently becoming increasingly significant due to the growing number of new WWTPs. Today we have at our disposal a great number of various technological possibilities for the treatment and disposal of sludge, and almost all of them are burdened with considerable initial-investment and plant-operation costs. One of the acceptable solutions involves the use of sludge and its by-products for various purposes (e.g. ash obtained by thermal treatment in concrete industry). From the aspect of sustainable development, the utilisation of sludge almost fully closes the waste water purification cycle, and the remaining negligible quantities of waste substances can be disposed into environment as waste. In the light of current worldwide trends, Croatia is also expected to place a greater emphasis on the reuse of sludge in the near future.

With regard to development of mechanical properties (compressive and bending strength) of cement mortars and/or concrete prepared with ash as a partial replacement of cement, two significant trends can be differentiated: the strength falls with and increase in the proportion of ash, while the strength increases with an increase in ash fineness. However, in general terms, strength values still remain somewhat lower compared to the strength exhibited by reference mixes.

Significant differences in the absolute values of results can be noted after comparison of results obtained by various authors. Such differences are primarily due to the difference in sludge (ash) properties, sample sizes, and w/c ratios used. The role of the ash generation process is also significant: type of furnace, temperature, influence of various additives added in the course of incineration, etc.

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