APPLICATION OF WOOD ASH AS A SUBSTITUTE FOR FLY ASH AND INVESTIGATION OF CONCRETE PROPERTIES

ANWENDUNG VON HOLZASCHE ALS ERSATZ FÜR FLUGASCHE UND UNTERSUCHUNG DER BETONEIGENSCHAFTEN

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SUMMARY

In the coming years, many German and European coal-fired power plants will be shut down in an effort to reduce the CO_2 emission. The shutdown will mean that fly ash, which is a by-product of the combustion process and has been a valuable additive in concrete for years, will no longer be available. Therefore, the aim of this work was to use wood ash as a substitute for fly ash and to investigate its influence on the concrete properties. Wood ash is a by-product of wood-fired combined heat and power plants, which primarily ends up as waste in landfills. The use of wood ash in the construction industry enables the recycling of wood ash and thus has a positive effect on the two aspects, namely environmental protection and economic efficiency.

An analysis of the chemical composition of the wood ashes showed that the cyclone ash meets the majority of the limit values from the fly ash standard. The requirements on heavy metals for soil and groundwater protection were all met. For testing the compressive strength, recipes with different ashes were tested and compared with the reference mixtures. From the results it can be seen that even higher compressive strengths are achieved with the mixtures with the cyclone ashes as additive than with mixtures with the fly ashes as additive. By using the wood ashes in the concrete, these concretes can be described as bio-economical wood ash concrete.

ZUSAMMENFASSUNG

In den kommenden Jahren werden viele deutsche und europäische Kohlekraftwerke aufgrund steigender CO_2-Emissionen abgeschaltet werden. Durch die Abschaltung wird Flugasche, die als Nebenprodukt des Verbrennungsprozesses anfällt und seit Jahren ein wertvoller Zusatzstoff im Beton ist, nicht mehr verfügbar sein. Ziel dieser Arbeit war es daher, Holzasche als Ersatz für Flugasche einzusetzen und ihren Einfluss auf die Betoneigenschaften zu untersuchen. Holzasche ist ein Nebenprodukt von holzbefeuerten Heizkraftwerken, das als Abfall auf Deponien landet. Die Verwendung von Holzasche in der Bauindustrie ermöglicht die Verwertung von Holzasche und wirkt sich somit positiv auf die beiden Aspekte Umweltschutz und Wirtschaftlichkeit aus.

Eine Analyse der chemischen Zusammensetzung der Holzaschen zeigte, dass die Zyklonasche die meisten Grenzwerte der Flugaschenorm einhält. Die Anforderungen an Schwermetalle für den Boden- und Grundwasserschutz wurden alle erfüllt. Zur Prüfung der Druckfestigkeit wurden Rezepturen mit unterschiedlichen Aschen durchgeführt und mit den Referenzmischungen verglichen. Aus den Ergebnissen ist ersichtlich, dass mit den Mischungen mit den Zyklonaschen als Additiv noch höhere Druckfestigkeiten erreicht werden als mit den Mischungen mit den Flugaschen als Additiv. Durch die Verwendung der Holzaschen im Beton können diese Betone als bio-ökonomischer Holzaschebeton bezeichnet werden.

1. INTRODUCTION

Future-oriented building is an important topic of our time. In order to realize a building project, building materials and methods of processing which allow for a cost-effective production are needed. For this reason, ecology and economy are very important aspects of future-oriented research. Ecological goals are energy and resource efficiency and the reduction of CO_2 emission.

In Germany, the bio-economy is a major topic with the main goal of linking sustainability with the economy. Possibilities are being explored to minimise the burden on the environment and replace fossil raw materials. A very important point in this topic is the reduction of CO_2 - emission. With regard to this point, wood is an optimal raw material, because wood is CO_2 - neutral and a renewable resource. Wood absorbs CO_2 during its growth phase and stores it. This means that wood has a CO_2 cycle which is balanced out during combustion. This means that burning wood is climate-neutral, as no additional CO_2 is produced, but the absorbed CO_2 is released. Wood is a renewable raw material which is excellently suited for the production of heat and energy. There are many power plants in Europa that make a major contribution to the production of electricity. Wood chips are used to generate electricity in these power plants. This in turn is waste that is produced in the manufacture of wood products, landscape conservation or as residual forest wood. Therefore, no additional trees need to be felled to generate electricity. The combustion of the wood chips produces three ash fractions. These are the grate ash, which is the only ash that has a permit for use as a fertilizer, the cyclone ash and the electrostatic precipitator ash. Cyclone ash, depending on the power plant, accounts for 30-60% of production, electrofilter ash for 5-15%. The rest are the coarsest (grate) ashes for 20-30%. The electrofilter ash has the highest fineness between the ash fractions, the particle size is smaller than 10 µm. In case of cyclone ash about 2 -160 µm.

One of the major problems with the wood combustion is the residual waste - wood ash. Large quantities of wood ash are stored as waste in landfills, which may lead to environmental pollution due to leakage of heavy metals in the ground water. Hence, it is necessary to find possibilities for recycling wood ash. With the tendency of ever reducing coal combustion, it can be expected that the combustion of wood will increase over the next decades, thus increasing the possible environmental impact of the wood ash. Now the question arises - how can these ashes be recycled and brought into the economic cycle or into the climate-neutral economic system of the bio-economy [1-2]?

In the construction industry, cyclone ash can be used as a secondary main component of cement or as an additive in concrete. Thus, the wood ashes could be recycled and thus have the advantage that landfills are not overloaded and the use of natural primary materials is reduced. The designation as waste for the secondary materials would be dropped under the Waste Management Act, since recyclable materials are classified as economic goods. However, in order to be used in concrete, the wood ashes must meet certain requirements. The main requirements are the strenght, durability and permanent integration of environmentally relevant elements, like heavy metals [3].

The present work focuses on the investigation of electrofilter and cyclon ash as possible additives in concrete. Grate ash is not considered, mainly due to its high organic content.

2. CONDUCTED INVESTIGATION

In order to be used in concrete, wooden ashes must meet the requirements regarding the chemical composition, fresh and hardened concrete properties as well as durability. As with all other additives, the harmful components in the wood ashes may only be contained in limited quantities.

The ashes examined in this work come from different tree species that were burned together. The filter ash is brighter than the cyclone ash [4-6]. In the context of this work chemical, physical and structural properties of the wood ash were investigated. For a better comparison, different reference materials were used in the present study: Portland cement CEM I 42.5, blast furnace slag cement CEM III 42.5 and fly ash from the power plant in Opole, Poland (certified for use in concrete).

The chemical elements of the wood ashes and fly ash were determined from the aqua regia digestion according to DIN EN 13657 [7] and the borate exclusion according to DIN ISO 14869-2 [8].

The proportions of heavy metals in the analyzed wood ashes are shown in Table 1 and compared with the requirement values for the solid content of waste for use in building products of the German Institute for Building Technology (DIBt) [9]. It can be seen that the cyclone ash completely complies with the limit values for heavy metals. However, the amount of cadmium and zinc in the electrofilter ash is clearly exceeded.

Table 1: Share of heavy metals in wood and fly ash

Element	**Unit**	**Electrofilter ash**	**Cyclone ash**	**Fly ash**	**Limit values acc. to DIBt**
Antimony Sb	mg/kg	52	6	3	-
Arsenic As	mg/kg	149	14.7	31.3	150
Cadmium Cd	mg/kg	36.2	4.7	1.2	< 10
Copper Cu	mg/kg	321	172	35	< 500
Molybdenum Mo	mg/kg	34	6	36	-
Nickel Ni	mg/kg	34	35	43	< 500
Mercury Hg	mg/kg	0.43	< 0.07	0.1	< 5
Selenium Se	mg/kg	9	1	14	-
Thallium Tl	mg/kg	6.9	< 0.2	1.6	7
Zinc Zn	mg/kg	8970	722	118	< 1500

Table 2 shows the chemical components of electrofilter ash, cyclone wood ash and coal fly ash. The limit values valid for fly ash according to EN 450 are also listed for better comparison. In principle, cyclone ash fulfills the majority of the normative requirements (limit values) for the coal fly ash, with a slightly lower content of Si-, Al- and Fe-oxides than required for fly ash. The results show that the amount of CaO is above the limit value for silica-rich fly ash.

In the case of electrofilter ash, the majority of the treshold values are not met [10-13]. The content of the reactive oxides is very low, indicating low pozzolanic potential. High contents of Na_20, SO_3 and Cl could lead to poor durability of concrete.

Table 2: Chemical composition of electrofilter ash, cyclone ash and fly ash

Element	Unit	Electrofilter ash	Cyclone ash	Fly ash	Limit values
SiO_2	M.-%	9.51	52.94	54.27	≥ 25
Al_2O_3	M.-%	2.32	8.70	19.60	-
Fe_2O_3	M.-%	1.52	3.41	11.83	-
SiO_2 + Al_2O_3 + Fe_2O_3	M.-%	13.35	65.05	85.70	≥ 70
TiO_2	M.-%	0.13	0.60	1.10	-
MnO	M.-%	0.27	0.18	0.06	-
MgO	M.-%	4.06	2.31	1.84	≤ 4
CaO	M.-%	30.67	17.76	2.48	≤ 10
K_2O	M.-%	14.10	5.86	3.81	-
Na_2O	M.-%	1.25	1.08	0.76	-
Na_2O_{Eq}	M.-%	10.53	4.94	3.27	≤ 5
P_2O_5	M.-%	1.62	0.72	0.47	≤ 5
SO_3	M.-%	14.55	1.00	0.28	≤ 3
Cl	M.-%	4.58	0.01	0.03	≤ 0.1
SrO	M.-%	0.07	0.04	0.08	-
ZnO	M.-%	1.09	0.10	-	-

In the next step, further chemical properties of the wood ash such as loss on ignition according to EN 196 [14] and physical properties such as specific surface according to EN 196 and DIN 1164 [15] were determined.

The loss on ignition describes the amount of organic particles that are not burnt at a high temperature of 900°C. These particles absorb more water during concrete production and can adversely affect the efectiveness of additives as well as the freeze-thaw resistence. Therefore, the higher the loss on ignition, the higher the water requirement. Based on the loss on ignition, fly ashes are typically divided into three categories in EN 450-1. Category A with the content of organic components ≤ 5 wt.%, category B ≤ 7 wt.% and category C ≤ 9 wt.%. In Germany, the use of fly ash as an additive is only permitted for category A [13]. Table 3 shows the results for the investigatet ashes. It can be seen that the cyclone ashes are in category A, so they can be used as a potential additive. Electrofilter ash on the other hand fulfils the requirements only for class C.

Table 3: Loss on ignition of electrofilter and cyclone ash

Sample	**m_1 [g]**	**m_2 [g]**	**Loss on ignition [wt.-%]**	**Mean value [wt.-%]**	**Category (EN 450-1)**
EA 1	1.019	0.939	7.85	8.13	C
EA 2	1.023	0.939	8.21		
EA 3	1.017	0.932	8.32		
ZA 1	1.006	0.969	3.68	3.79	A
ZA 2	1.025	0.988	3.61		
ZA 3	1.008	0.967	4.07		

3. PHYSICAL PROPERTIES OF THE INVESTIGATED MATERIALS

The specific surface of the ashes was determined by laser granulometry. This method is used to determine the specific surface of very fine materials, such as microsilica. This gives more meaningful results for cyclone ash than the Blaine method. The bulk density for the coal fly ash is about 2.25 g/cm^3 and the specific surface is about 3000 cm^2/g. Table 4 shows that the bulk density (ρ) for both ashes is very close to the values of the fly ashes, but the specific surface area of the eleftrofilter ash is much higher than those of coal ash and cyclone ash. These are in the range of the specific surface of microsilica, which typically lies between 15000 and 35000 cm^2/g. [6].

The fineness is determined by air jet sieving according to EN 451-2. Reference sieve with a mesh size of 0.045 mm is used to sift the sample. To carry out the test, the samples are first dried in an oven at (105 ± 5) °C. Here 10 g per sample and per test is used. The result of the tests is reported as the residue on the 0.045 mm sieve. The upper limit value for fly ash is 40% residue on 0.045 mm sieve [16]. The results of the sieving for the investigated ashes are shown in Table 4. The cyclone ash has an average value of 35.3% and is thus below the limit value determined for fly ash (≤ 40 wt.% category N). Due to very small particle size, it was not possible to measure the fineness of electrofilter ash using this method.

Table 4: Specific surface area and density for electrofilter and cyclone ash

	Electrofilter ash		**Cyclone ash**	
Bulk density ρ [g/cm^3]	2.48	2.48	2.55	2.55
Specific surface area S [cm^2/g]	25799	25805	2635	2648
Mean value of the Specific surface area S [cm^2/g]	25800		2640	
Fineness according to EN 451-2 (wt.%)	not measurable		35.3	

4. INVESTIGATION OF THE HARDENED CEMENT PASTE

The compressive and flexural strength are tested on pure hardened cement paste. The compound of water and cement is called cement paste in its fresh state and hardened cement paste in its solid state. For the compression tests, cement pastes with different proportions of cement and additives are produced. This allows the development of compressive strength to be investigated as a function of the amount of additive. The standard cements CEM I and CEM III are used for the production of the samples. The Portland cement CEM I 42,5 N was chosen to show the difference between a cement with the main component cement clinker and the replacement of the cement clinker by the hard coal fly ash and wood ash. The blast furnace cement CEM III 42,5 N is used to test the compressive strength resulting from the interaction of the secondary main component of the cement and the ashes. Thus, a mixture is obtained whose clinker content is additionally reduced with the clinker-reduced cement CEM III through the use of additives. The dimensions of the test specimens are 40 mm x 40 mm x 160 mm, as specified in EN 196-1 [17].

Reference cement paste with pure Portland cement CEM I 42.5 N (abbreviation RB) was used as reference material. Subsequently, the cement content was replaced by 20 wt.% with fly ash (FA) and cyclone ash (ZA), respectively. In order to be able to investigate the effect of fly ash and wood ash, a further mixture was produced from their combination. The proportion of fly ash was 10 wt.% and of cyclone ash 10 wt.%. The same procedure was done with the electrofilter ashes (Fig. 1). All values of ashes have fulfilled the normative requirements for 75% of the compressive strength after 28 days of reference paste (requirements for fly ash acc. to EN 450).

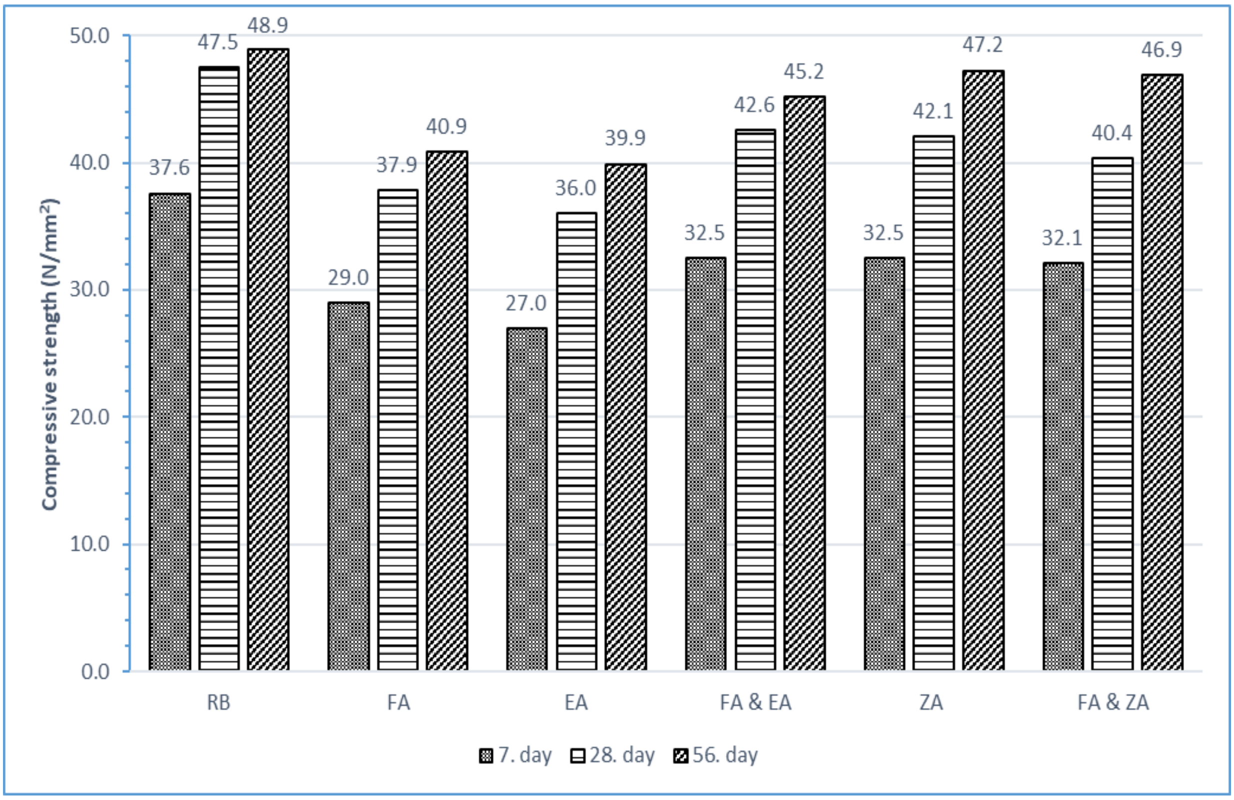

Fig. 1: Compressive strength of reference cement paste (RB), fly ash (FA), electofilter wood ash (EA), combination (FA & EA), cyclone wood ash (ZA) and combination (FA & ZA), at 20 wt.% cement substitute and 0.5 w/c value after 7, 28 and 56 days

Fig. 1 shows that cyclone ash (ZA) and the combination of fly ash and cyclone ash (FA & ZA) have higher compressive strength than pure fly ash. The cyclone ash alone, with a w/c ratio of 0.5 and 20 wt.% cement substitute, has a compressive strength comparable to that of the reference mix (RB) after 56 days. The electrofilter ash (EA) has a somewhat lower strength than the cyclone ash (ZA). This is also expected considering the chemical composition of the two ashes (low content of reactive oxides in the electrofilter ash). It is presumed that the contribution of the electrofilter ash toward the compressive strength is to a large extent due to the fineness of the material, which resulted in an improved filler effect.

Fig. 2 shows the results for mixing with cyclone ash in combination with blastfurnace cement and with Portland cement. The compressive strength with CEM III at a w/c value of 0.50 and a cement substitute of 20 wt.% is significantly higher than with CEM I for both types of ash. This has the advantage of a higher clinker reduction, since CEM III is already a clinker-reduced cement. As a result, the environmental impact is reduced and an even more ecological concrete is produced compared to CEM I in combination with wood ash.

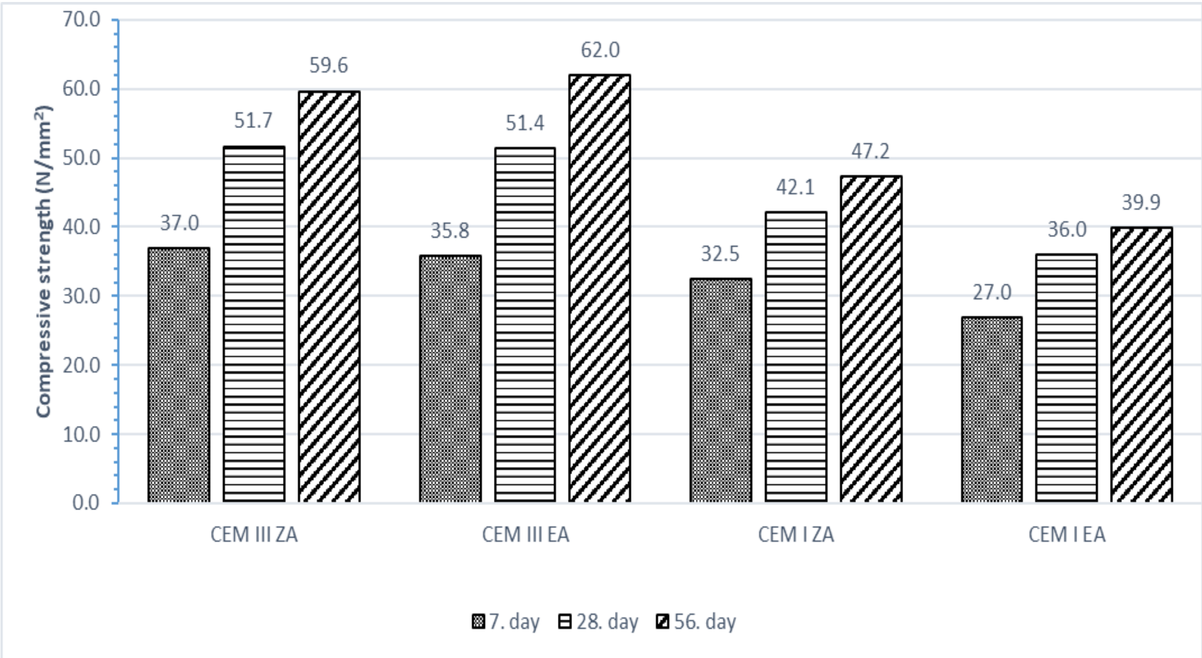

Fig. 2: Comparison of the compressive strength of electrofilter and cyclone ash with CEM I and CEM III at 20 wt.% cement substitute and 0.5 w/c value after 7, 28 and 56 days

5. INVESTIGATION OF CONCRETE WITH ELECTROFILTER WOOD ASH

In this work, the use of wood ash is carried out in normal concrete in order to test the compressive strength for concrete. For the hardened concrete tests, cubes according to EN 1045-2 with an edge length of 150 mm are produced. The cubes were demolded after 24 h and stored at a temperature of (20 ± 2) °C for 6 days in a water bath or humidity chamber with a relative humidity of more than 95%. Afterwards, the cubes were stored at a room temperature of (20 ± 2) °C and a relative humidity of (65 ± 5) % until the test day [18].

The formulations for the normal concrete were calculated with a water-cement ratio of 0.40. The quantities of superplasticizer were calibarated beforehand to assure good workability. The reference concrete (RB) is the reference concrete, which contains no additives and a superplasticizer quantity of 5.04 kg/dm^3 (Table 5). Despite a stiffer consistency, the mixture could be compacted in the moulds on the vibrating table. The fly ash concrete (FA) contains fly ash, which replaces 25% of the cement by weight. The amount of superplasticizer is lowest here at 1.68 kg/dm^3 and allows easy compaction on the vibrating table. The electrofilter ash concrete (EA) contains electrofilter ash instead of fly ash, which also replaces 25% of the cement by weight. Due to the technical difficulties in procuring the material, it was not possible to perform tests on concrete with cyclone ash.

The amount of superplasticizer is the same as for the reference concrete. However, here the consistency was much stiffer and a fast and forceful compaction was necessary. A higher amount of superplasticizer would facilitate the processing. For the concrete (RB) and (FA), the specimens were removed from formwork after 24 hours. The normal concrete (EA) sets much faster, but hardens slower than the normal concrete (FA). For this reason, it could only be demolded after 48 hours.

Table 5: Normal concrete formulations

Starting materials	**Unit**	**Normal concrete Quantity [kg/dm^3]**		
		RB	**FA**	**EA**
Cement CEM I 42,5 R	kg/dm^3	420	315	315
Fly ash FA	kg/dm^3	-	105	
Electrofilter ash EA	kg/dm^3	-	-	105
Water	kg/dm^3	168	168	168
Aggregate 0/4	kg/dm^3	536.5	536.5	536.5
Aggregate 5/8	kg/dm^3	1283	1283	1283
Superplasticizer	kg/dm^3	5.04	1.68	5.04

For the consistency of the fresh concrete, the flow spread is determined according to DIN EN 12350-5 [19]. The results are presented in Table 6.

Table 6: Consistency of the investigated concrete

Normal concrete	**Diameter d_1 [cm]**	**Diameter d_2 [cm]**	**Flow spread [cm]**	**Class**
RB	57	58	57.5	F5
FA	64	64	64	F6
EA	50	54	52	F4

The compressive strength tests are carried out after 7 days and after 28 days in accordance with EN 206 [20]. The compressive strength of normal concrete (RB) has already reached a very high compressive strength on the 7th day, see Fig. 3. On the 28th day, there was an increase in strength of approx. 15%. For normal concrete (FA) and (EA), the increase in strength from the 7th to the 28th day is approx. 30%.

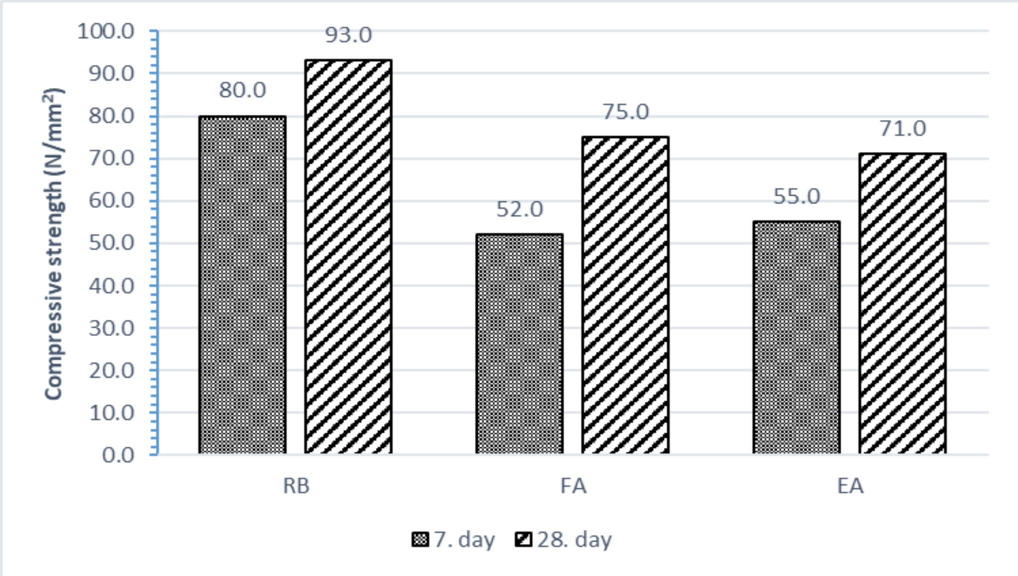

Fig. 3: Compressive strength of normal concrete RA, FA and EA after 7 and 28 days

The determination of pore size distribution and porosity is carried out according to DIN 66133 [21] with mercury pressure porosimetry. Prisms made of the three different normal concretes (RB), (FA) and (EA) are prepared for this test. After 28 days of storage, these are kept in an oven at 105°C for three days. Thus, the water contained in the concrete evaporates and further hydration is prevented. After the water has evaporated, the prisms are crushed into approx. 5 mm pieces. The aggregates are removed from the hardened cement paste, so that the pure hardened cement paste is available for the tests.

The pore volume distribution and specific surface area can be determined as a result of mercury pressed into the pores under pressure. The use of mercury is important because it is a non-wetting liquid. The experiments are performed with the two instruments Pascal 140 and Pascal 440 from Thermo Scientific. For this purpose, 2 to 2.5 g of the prepared samples are filled into penetrometers and weighed, the penetrometer is placed in the 1st instrument Pascal 140, which carries out the measurement of the macro-pore distribution. Here the air is extracted from the sample by vacuum. Then mercury is filled in and pressed into the pores with gradually increasing pressure. The total pore volume can be calculated from the amount of mercury filled into the pores. After this measurement, the Penetrometer is put into the 2nd device Pascal 440. This device determines the macro- and mesopore distribution. For this purpose, the penetrometer in the device is filled with oil so that no air is contained. The graphical representation of the pore size distribution is derived from the injected mercury volume in relation to the mass of the sample as a function of the pore radius [21].

The different size ranges of the pores are divided as follows. Pore widths between 10^0 and 10^{-2} mm are called macropores. Smaller pores than these are called capillary pores and are in the range of 10^{-5} to 10^{-2} mm. The smallest pores are the micropores, which are in the range of 10^{-6} and 10^{-5} mm.

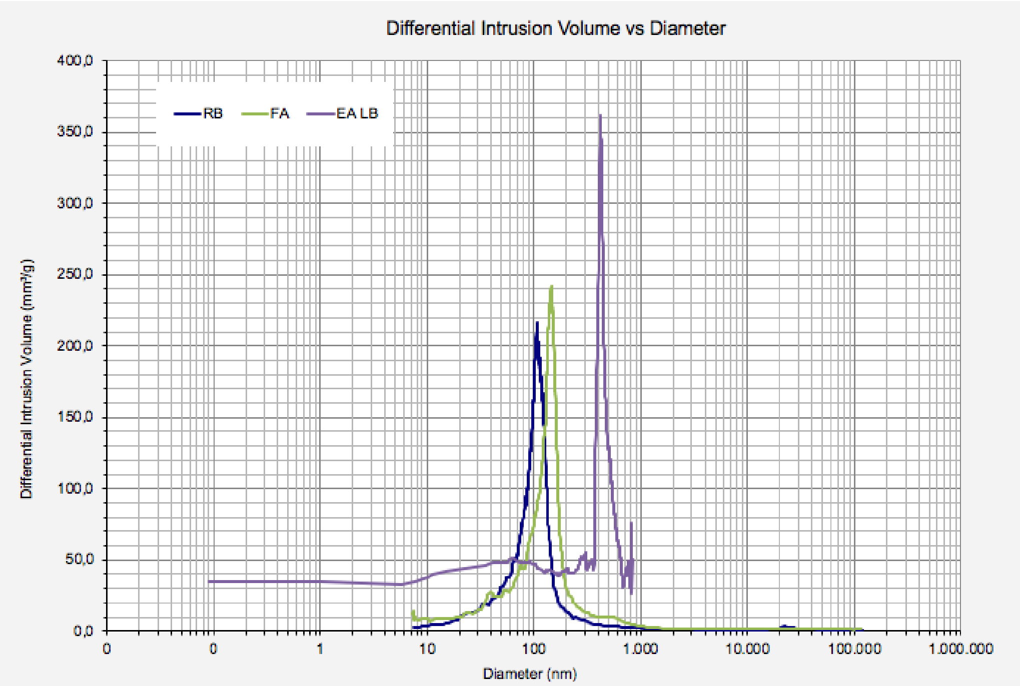

Fig. 4: Logarithmic display of the results

The results of mercury pressure porosimetry are shown in Fig. 4 and Table 7. The total porosity of the investigated concretes is very similar and are in the range of 14 to 15%. The capillary pore content of RB and FA is approx. 89.0% and of EA 60.6%. In comparison, the macro and micro pore content is very low in the reference concrete RB and fly ash concrete FA. The macropores are below 7% and the micropores below 5%. The electrofilter ash concrete EA, like the other two concretes, has a low micropore content, but a considerably higher macropore content of 36.2%. Concrete EA also shows a clear difference to concretes RB and FA in the ratio of capillary pores to macropores. The pore diameter r_{max} at the maximum of the differential size distribution curve is in the capillary pore range for all concretes. The volume of mercury VHG extruded is between 64.27 and 74.31 mm^3/g. The specific surface area is highest for EA at 4.859 m^2/g.

Table 7: Results of mercury pressure porosimetry

	RB	**FA**	**EA**
Porosity [%]	14.16	15.78	15.29
Intruded mercury volumes V_{HG} [mm^3/g]	64.27	74.31	70.87
Specific surface [m^2/g]	3.562	4.068	4.859
Macropores [%]	6.90	5.95	36.19
Capillary pore content [%]	89.75	89.18	60.59
Micropore content [%]	3.35	4.87	3.22
Ratio capillary pores/macropores	1:0.08	1:0.07	1:0.6

6. AVAILABILITY OF WOOD ASH

Every year millions of tonnes of ashes of different fractions are produced throughout Europe with an increasing tendency. In the years between 2010 and 2015, 75 million tonnes more wood were burnt in the EU than in the previous period (2005-2010) [22]. Many countries, such as Belgium, Denmark, the Netherlands and the UK, have built new wood-fired power plants or converted coal-fired ones so that they can now produce electricity from wood combustion. The Drax power station, the largest power station in the Kingdom, supplies seven percent of the country's electricity, with 70 percent of its electricity obtained from pressed wood pellets instead of coal. Around one million tons of wood ash is produced each year in Germany [23]. It is expected that these numbers will further rise in future. Considering that the yearly consumption of fly ash in Germany lies slightly below 3 million tons, the available amount of wood ash can at least partly compensate for the possible shortage of fly ash in future.

7. SUMMARY

In the present work, preliminary results on the performance of two different fractions of wood ash as a possible cement additive are reported and discussed.

The analysis of the material properties shows that the composition and propreties of the cyclone ash are very close to the values of the fly ash. The limit values regarding the chemical composition and heavy metals content on the fly ash according to EN 450-1 are largely met by the cyclone wood ash. On the other hand, electrofilter ash contains a very low content of reactive oxides and relatively high content of possibly harmful components for use in concrete, such as heavy metals, alkalies and sulfates.

From the results of the compressive strengths of the hardened cement paste, it is clear that the replacement of cement by cyclone ash provides higher compressive strengths than fly ash. Here the compressive strengths are almost identical to the results of the reference mixture (pure Portland cement). Similar results are also obtained when using blast furnace cement CEM III instead of pure Portland cement. It is interesting to observe that the electrofilter ash with relatively low content of reactive oxides also developed relatively high strength. This may be partially due to the very high fineness of the material, which enhanced the filler effect.

For the future it is necessary to check the requirements of durability, including resistance to chemical attack, freeze-thaw resistance and corrosion protection of the reinforcement. Cyclone ash, like coal fly ash, is a type II additive, as it reacts with water and cement. The use of wood ashes has many economic and ecological aspects. The wood ashes that are nowadays considered as waste can be used as a valuable cement and concrete additive, furthermore the space on the landfill can be gained. For ecological reasons, the use of wood ash in cement or concrete significantly improves the eco- balance. Wood ash replaces cement clinker, which emits an enormous amount of CO_2 during the production process. Wood ashes can be an optimal solution for the disappearing coal fly ash in the future.

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