EVALUATION OF WOOD FLY ASH AS A CEMENT REPLACEMENT MATERIAL IN CONCRETE

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Abstract
Biomass is the largest source of renewable energy in the European Union, of which 60% comes directly or indirectly from forests. The most common way of producing energy from biomass is combustion, and this process yields substantial quantity of ash. The cement and concrete industry has been identified as one of the main potentials for biomass ash utilization. This paper presents the results of an experimental study aimed at determining the influence of wood fly ash on the properties of concrete. Wood ash was sampled from three power plants using different technologies of incineration and different types and parts of wood were used as a fuel which consequently had a large impact on the properties of ash. Subsequently, workability, heat of hydration, stiffness development, 28-day compressive strength, apparent porosity, and capillary absorption were determined on concrete mixes prepared with WFA as cement replacement from 5–45% by weight. Cement replacement up to 15% with the finest WFA accelerated hydration, stiffness development, and increased compressive strength of concrete up to 18%, while replacement with coarser WFA's led to a decrease in compressive strength of up to 5% and had more gradual heat liberation. The dominant effect that could explain these findings is attributed to the filler and filling effect mechanisms.

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Keywords: biomass; wood fly ash; supplementary cementitious materials

1. Introduction

The need to reduce greenhouse gas emissions accelerates the transition to renewable energy sources. Biomass is the largest source of renewable energy in the European Union (EU), of which 60% comes directly or indirectly from forests [1, 2]. The most common way of producing energy from biomass is combustion, and this process yields on average a quantity of ash between 2.7% and 3.5% of the original weight of wood biomass [3]. It is estimated that in 2018 about 11 million tons of ash were produced in the EU-28 countries, most of which is wood biomass ash (WBA) [4].

During combustion in a biomass plant, two types of ash are generated: bottom ash and fly ash. Ash from the combustion of natural woody biomass contains valuable plant nutrients such as K, P, Mg, and Ca, most of which are contained in the bottom and coarse fly ash, while volatile heavy metals are concentrated in the fine fly ash fraction [5]. Therefore, it has been suggested that the bottom and coarse fly ash fraction should be returned to the forest from which they originated, while the fine fly ash should be utilized in industrial processes or disposed of [5].

The cement and concrete industry has been identified as one of the main potentials for biomass ash utilization [6]. The application of WFA is currently outside the scope of the standard for fly ash for concrete (EN 450-1:2012). Extensive research has been conducted to investigate the possibilities of using WFA as a cement re-placement material [7–16]. It has been reported that WFA from grate combustors and fluidized bed combustors can have hydraulic and/or pozzolanic properties [7, 10, 13]. The utilization of WFA as a cement replacement modifies the workability and mechanical properties of the tested material. As the cement replacement level increases, water demand usually increases and the compressive strength decreases [7–12, 14]. However, replacing cement with WFA can also increase compressive strength when the cement replacement level is low [11, 17]. Improved workability has also been found when WFA was used as a filler and partial replacement for fine aggregate in concrete [13].
Most of the studies on the influence of WFA produced during combustion in biomass plants on the properties of cement composites have been tested on cement pastes and mortars and only a small number of experiments have been scaled up to the concrete level. To develop guidelines for the use of WFA in structural concrete, the interdependence between properties of WFA and the properties of fresh and hardened concrete must be clearly established. The aim of the experimental work presented here is to show the influence of WFA with different physical and chemical properties, used as partial cement replacements, on the properties of fresh and hardened concrete and to identify the most probable mechanisms that govern these changes. The WFA used originates from three different power plants with two types of incineration technologies.

2. Materials and methods

2.1. Characterisation of fly ash and cement

Wood fly ash (WFA) is collected from three powerplants in Croatia. All three plants are co-generation biomass plants producing both heat and electrical energy. Plant F4 is the smallest plant with 1 MW of electrical capacity and 4.1 MW of heat capacity. Wood species mostly used as a fuel are given in Table 1 and the parts of wood used as a fuel consist of wood chips made from roundwood and thinning residues, including twigs, tops, and branches. Plant F5 has a production capacity of 2.75 MW of electrical and 15 MW of heat energy. Plants F4 and F5 use fixed bed combustion in which wood fuel is burned in a grate furnace [18]. Plant F6 is the largest biomass plant with the capacity of production of 8.6 MW of electrical and 16 MW of heat energy. Wood fuel is used in the form of wood chips made from whole trees containing bark, twigs, and leaves. Plant F6 uses a bubbling fluidized bed combustion system with quartz sand as a bed material [18]. The fly ash particles carried from the combustor are captured by bag filters [18].

The information about the incineration temperature of wood biomass is given by the technologists in the power plant. The chemical and physical properties of WFA used in this work are presented in Table 1. The median particle size (d50) is determined from a laser diffraction method using a dry measurement. Elemental composition is determined by X-ray fluorescence according to standard ISO/TS 16996:2015. Loss on ignition (LOI) has been determined according to ASTM D7348-13, density according to ASTM C188-17, and pH value according to EN 12176:2005. More detailed information about fly ash characterisation is presented in [4].

2.2. Concrete mix design and testing methods

WFA was stored in the laboratory for a period of approximately six months before being used in concrete. During storage, WFA was kept sealed in plastic bags and then stored in closed plastic containers. The cement used in this study was Portland cement type CEM I 42.5 R conforming to European standard EN 197-1:2011. The aggregate was crushed dolomite with an average bulk density of 2.8 kg/dm³ and absorption of 0.47%, 0.66%, and 0.32% for fractions 0/4, 4/8, and 8/16 mm, respectively.

The compositions of the concrete mixes are given in Table 2. Each mix is designated according to the WFA used and the cement replacement percentage. All mixes had the same water/(cement + WFA) ratio of 0.5. A total of 7 concrete mixes containing WFA were prepared with cement replacement percentages of 15% and 30% for ash F4, 15%, 30% and 45% for ash F5, and 5% and 15% for ash F6. In the first stage of the experimental work, a reference concrete mix (M0) and mixes with 15% cement replacement were made. The decision to increase or decrease the cement replacement level in the further mix design process was based on the influence of WFA on the workability of the fresh concrete. Workability was evaluated by the slump test, and cohesiveness was determined by visual inspection of each mix. Before mixing, all constituents were conditioned to a temperature of 20 ± 2°C.

In the fresh concrete consistency by slump test (EN 12350-2:2019), density (EN 12350-6:2019), air content (EN 12350-7:2019), and temperature were measured. The concrete was compacted on a vibrating table. After compaction, the specimens were stored in a room with a temperature of 20 ± 5°C.
and covered with a plastic sheet. After 24 h, the specimens were demoulded and moved to a curing room where they were cured in the air at a temperature of 20 ± 2°C and a relative humidity of >95%. All measurements were performed on concrete from the same batch.

Table 1. Chemical and physical composition of cement and WFA

<table>
<thead>
<tr>
<th></th>
<th>CEM</th>
<th>F4</th>
<th>F5</th>
<th>F6</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>GC</td>
<td>GC</td>
<td>BFB</td>
<td></td>
</tr>
<tr>
<td>Combustion technology</td>
<td>-</td>
<td>700–950</td>
<td>up to 800</td>
<td>up to 850</td>
</tr>
<tr>
<td>Incineration temperature (°C)</td>
<td>beech, oak, fir, spruce</td>
<td>beech, oak, hornbeam</td>
<td>beech, oak, hornbeam, poplar</td>
<td></td>
</tr>
<tr>
<td>Wood type</td>
<td>P₂O₅</td>
<td>0.22</td>
<td>1.82</td>
<td>1.35</td>
</tr>
<tr>
<td></td>
<td>Na₂O</td>
<td>0.85</td>
<td>0.65</td>
<td>1.32</td>
</tr>
<tr>
<td></td>
<td>K₂O</td>
<td>1.25</td>
<td>6.05</td>
<td>4.77</td>
</tr>
<tr>
<td></td>
<td>CaO</td>
<td>59.80</td>
<td>46.75</td>
<td>16.25</td>
</tr>
<tr>
<td></td>
<td>MgO</td>
<td>2.01</td>
<td>8.26</td>
<td>4.30</td>
</tr>
<tr>
<td></td>
<td>Al₂O₃</td>
<td>4.94</td>
<td>6.16</td>
<td>10.50</td>
</tr>
<tr>
<td></td>
<td>TiO₂</td>
<td>0.23</td>
<td>0.34</td>
<td>1.17</td>
</tr>
<tr>
<td></td>
<td>Fe₂O₃</td>
<td>3.15</td>
<td>2.85</td>
<td>4.23</td>
</tr>
<tr>
<td></td>
<td>SiO₂</td>
<td>21.88</td>
<td>19.80</td>
<td>39.95</td>
</tr>
<tr>
<td></td>
<td>SO₃</td>
<td>3.33</td>
<td>2.73</td>
<td>0.60</td>
</tr>
<tr>
<td></td>
<td>LOI (at 950 °C)</td>
<td>3.60</td>
<td>3.80</td>
<td>8.30</td>
</tr>
<tr>
<td></td>
<td>pH</td>
<td>12.86</td>
<td>13.15</td>
<td>12.97</td>
</tr>
<tr>
<td></td>
<td>d₀ (µm)</td>
<td>9.4</td>
<td>71.9</td>
<td>120.7</td>
</tr>
<tr>
<td></td>
<td>Density (g/cm³)</td>
<td>3.10</td>
<td>2.59</td>
<td>2.63</td>
</tr>
</tbody>
</table>

GC – Grate combustor; BFB – Bubbling fluidized bed

Table 2. Concrete mix composition (quantities per 1 m³ of concrete).

<table>
<thead>
<tr>
<th>Mix Designation</th>
<th>M0</th>
<th>F4-15</th>
<th>F4-30</th>
<th>F5-15</th>
<th>F5-30</th>
<th>F5-45</th>
<th>F6-5</th>
<th>F6-15</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cement (kg)</td>
<td>380</td>
<td>323</td>
<td>266</td>
<td>323</td>
<td>266</td>
<td>209</td>
<td>361</td>
<td>323</td>
</tr>
<tr>
<td>WFA content (kg)</td>
<td>0</td>
<td>57</td>
<td>114</td>
<td>57</td>
<td>114</td>
<td>171</td>
<td>19</td>
<td>57</td>
</tr>
<tr>
<td>Cement + WFA (kg)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>380</td>
<td></td>
</tr>
<tr>
<td>w/(cem. + WFA) ratio</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>Water (kg)</td>
<td>190</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Aggregate (kg)</td>
<td>1821</td>
<td>1811</td>
<td>1801</td>
<td>1811</td>
<td>1801</td>
<td>1791</td>
<td>1816</td>
<td>1805</td>
</tr>
<tr>
<td>Fine aggregate (kg)</td>
<td>648</td>
<td>645</td>
<td>641</td>
<td>645</td>
<td>641</td>
<td>638</td>
<td>646</td>
<td>643</td>
</tr>
<tr>
<td>Coarse aggregate (kg)</td>
<td>1173</td>
<td>1167</td>
<td>1160</td>
<td>1167</td>
<td>1160</td>
<td>1154</td>
<td>1169</td>
<td>1162</td>
</tr>
</tbody>
</table>

The heat of hydration is monitored with the ToniCAL 7336 heat flow differential calorimeter on a cylindrical specimen with a diameter of 150 mm and a height of 300 mm. The rate of heat generation was monitored during the first 5 days of hydration.

Concrete stiffness development was monitored by measuring ultrasonic pulse velocity (UPV) at 1, 2, 7, and 28 days of age using a portable ultrasonic instrument with 54 kHz longitudinal wave transducers. The measurement was performed on concrete cubes with a side length of 150 mm. The same cubes were used for testing the compressive strength of concrete at 28 days of age according to standardized procedure (EN 12390-3:2019).

The capillary absorption measurement was performed on cylinder specimens 150 mm in diameter and height 50 mm, obtained by sawing from the standard cylinder 150 mm in diameter and height 300 mm. Upper and bottom slices of the cylinder were not tested to avoid effects of different boundary conditions. The first 10 mm of side surface in contact with water was coated with epoxy resin. Prior to testing, the
specimens were oven-dried at a temperature of 105 ± 5°C until the change in mass for two consecutive weighing became less than 0.5 g (≈0.025% of mass). After cooling to ambient temperature, the specimens were placed in a water container on cylindrical rods, and the water level was adjusted so that the bottom surface of the specimens was immersed in the water 2-5 mm. The mass of the specimens was measured at an interval of 5, 15, 30, 60, 120, 240, and 1500 min. From each mix, three specimens were tested for capillary absorption.

After the capillary absorption test, the water level in a water container was gradually increased at the rate of approximately 1/4 of the height of the specimens per day until the specimens were completely immersed. The specimens were kept under water until the mass of the specimens changed by less than 0.5 g after two consecutive measurements. The specimens were then weighted in surface dry conditions in air and in water. Bulk density in dry ($\rho_{z,dry}$) conditions, apparent solid density ($\rho_a$), and apparent porosity ($p_a$) were calculated according to Eqs. (1)-(3). The term apparent is used here because it is assumed that only open pores are filled with water.

$$
\rho_{z,dry} = \frac{m_{dry}\rho_w}{m_{sat} - m_{sat,w}},
$$

(1)

$$
\rho_a = \frac{m_{dry}\rho_w}{m_{dry} - m_{sat,w}},
$$

(2)

$$
p_a = \frac{m_{sat} - m_{dry}}{m_{sat} - m_{sat,w}} \cdot 100,
$$

(3)

where $m_{dry}$ is the mass of dry material, $m_{sat}$ is the mass of saturated material, $m_{sat,w}$ is the mass of saturated material weighted in water, and $\rho_w$ is the density of water.

3. Results and Discussion

The fresh concrete density varied between mixes in the range of 2440-2500 kg/m$^3$. For ashes F4 and F5, the density slightly decreased with increasing WFA content, while for ash F6, the density increased with increasing ash content. This observation on the fresh concrete was confirmed by the results of the bulk density measurements on the hardened concrete (Table 3). The air content was low in all mixtures, indicating dense packing of the concrete constituents after compaction on the vibrating table. The highest air content in mix F6-15 may be partially attributed to the low slump value, which made the compaction of concrete harder. The initial temperature of the concrete varied between 22.2 and 25.1 °C. Only mixes with ash F4 showed a consistent increase in temperature with the increase in ash content. This could be caused by fast initial heat liberation on contact with water but could also be attributed to temperature variations of the concrete constituents prior to mixing.

All mixes had adequate workability to be placed in moulds and compacted on a vibrating table without the loss of homogeneity. The influence of cement replacement on workability, as determined by the slump test, is shown in Table 3. Ash F6 at 15% cement replacement had the largest influence on slump values, where slump decreased from 90 mm measured on the reference concrete to 5 mm. Concrete produced with 15% cement replacement with F4 and F5 ashes showed only minor deviations from the slump value measured on the reference mix. A significant decrease in slump was also present with the F4-30 mix. The cement mix with ash F5 had almost no influence on the slump values. However, mix F5-45 gave a harsh concrete typical for low cement content concrete [21]. This mix also showed increased bleeding and a tendency for the segregation of the largest aggregate particles. It is also interesting to note that the F4-15 and F6-5 mixes showed an increased slump compared to the reference mix. This increase in slump is within the reproducibility limits for the test method (EN 12350-2:2019) but may also reflect the net effect caused by the cement replacement with WFA.
SEM images (presented in [4]) show that all the WFAs used in this work contain both irregular and spherical particles and the main difference is in the size of the particles. Compared to the WFA particles, the cement particles seem to be more irregular, which is to be expected since these particles were formed by crushing larger clinker grains. Therefore, replacing cement with WFA increases the “sphericity” of the particles. Another important effect on workability is the water absorbed by the porous particles. The main content of the LOI in fly ash is unburned carbon, which has a high porosity and a very large specific surface and can absorb a significant amount of water [22]. Ash F6 contained particle sizes in a range very similar to Portland cement. The addition of ash F6 to the concrete increases the sphericity of the particles, which, in combination with the small particle sizes improved the packing and enhanced the workability.

Ash F6 also contained the highest amount of unburned carbon (LOI 12.7%), which absorbed a certain quantity of water. The increase in slump in mix F6-5 could be attributed to the enhanced workability due to the filling effect, while at 15% replacement the effect of water absorption and enhanced cohesiveness become dominant. The F4 ash contained coarser particles than the Portland cement, but it also contained 30% of particles <45 µm. This quantity of fine particles together with the increased circularity of the particles enhanced fluidity. At the same time, the portion of particles >45 µm loosens the particle packing, leading to increased water demand. The increase in slump in mix F4-15 could be attributed to the reduced water demand due to the filling effect, while at 30% cement replacement, the loosening of particle packing becomes dominant. Ash F5 contains only 7% of particles <45 µm, so it does not have capability to reduce the water demand through the filling effect. A loose particle packing is created, which decreased the cohesiveness of the mix, while part of the water was probably absorbed by the unburned carbon particles (LOI 8.3%). The existence of optimum WFA content in terms of concrete workability has been observed in [21, 23]

### 3.1. Heat Generation

Measurements of heat output started 30–90 min after initial contact of cement and water due to the time needed to perform tests on fresh concrete and, if necessary, the additional time required to precondition the specimen so that the concrete temperature is as close as possible to reference temperature of 23°C. All the heat flow curves in Fig. 1 have one significant peak separating the acceleration and deceleration periods of heat release. Replacement of cement with F4 and F5 ashes decreased the peak value of heat flow. Furthermore, the higher the ash content, the larger reduction of heat generation is. At a 15% replacement level, these ashes had almost the same effect on the heat flow. The difference in heat flow between the mixtures with F4 and F5 ashes appeared at a 30% replacement level, and in the mixtures made with F5 ash, there is a larger delay in the peak heat flow and the slope of the curve decreases. Increasing the replacement level to 45% further delayed the hydration process. Effects on heat flow similar to those of ash F5 have been reported for mixtures of Portland cement and type C coal fly ash and have been attributed to the disruption of the aluminate-sulfate balance in mixtures containing >20%
fly ash [24]. Roszczynialski and Nocun-Wczelik [25] also reported that low SO$_3$ content relative to aluminates produced a significant peak after 13 h caused by aluminates. Ash F5 has a significantly higher ratio of Al$_2$O$_3$ to SO$_3$ than cement or both ash F4 and F6 (Table 1), so this could be the main cause of the changes in the course of early hydration in mixtures containing ash F5.

Contrary to mixtures containing ash F4 and F5, mixtures containing ash F6 in both replacement levels had an increased rate of heat liberation compared to the reference mixture. It is well known that even inert mineral admixtures, when blended with cement, can accelerate hydration through filler effect [26]. The increased cohesiveness of the fresh concrete, the reduced porosity (Table 3), and the increased hydration rates found in mixtures with ash F6 (Fig. 1) can all be related to the filler effect.

3.2. Compressive strength

Compressive strength was determined on four specimens from each mix. The average 28-day compressive strength and standard deviation are given in Fig. 2. For both ash F4 and F5, the compressive strength decreased with the increase of ash content in the concrete but for the mixtures with ash F6, the compressive strength increased with the increase of ash content. As already mentioned, the replacement of cement with ash F6 resulted in the densest particle packing which is the main reason for the increase in compressive strength.

The reduction in compressive strength in WFA concrete may also be related to decreased aggregate content and the simultaneous incorporation of ash particles with lower stiffness and strength into the cement matrix. The density of WFA is lower than the density of cement, so the same mass of WFA occupies a larger volume than cement. This results in a reduction in the volume of aggregates in mixtures containing WFA (see Table 2). Since aggregates have greater strength and stiffness than WFA, their replacement by WFA weakens the concrete structure [27]. The decrease in strength may also be related to the lower content of cementitious material when part of the cement is replaced by WFA, as its reactivity is lower than that of the cement.
3.3. Stiffness development

During the first two days, the mixtures with 15% cement replaced by ash F4 and F5 exhibited slower stiffness development compared to the mixture M0 (Fig. 3). Mixtures containing ash F6 showed an increased rate of heat generation so the accelerated stiffness development can be attributed to an improved hydration process (Fig. 3).

Mixtures made with ash F4 and F5 exhibited a decrease in heat generation rate so that fewer hydration products were formed within the first days of hydration and the stiffness development is slowed down accordingly. Measurement over the 2–28-day period indicate that the addition of WFA changed the course of stiffness development. In the reference mixture, the period of 2–7 days shows the largest increase in UPV and a very low increase in UPV from 7–28 days. In mixtures with WFA, the increase in UPV is two to five times larger in the period of 7–28 days compared to the reference mixture. This could be due to the reactivity of ash with the species present in the pore solution.
3.4. Capillary absorption

The capillary absorption coefficient for each mixture was calculated and presented in Fig. 4. The capillary absorption coefficient was calculated using the average slope of the water intake curve over the period 120–1500 min. The initial 120 min of measurement was excluded from regression analysis to reduce the impact of nonlinearities contained in the first period of measurement on the value of the coefficient of capillary absorption. The capillary absorption coefficient and its standard deviation show that the replacement of cement with ash F4 and F5 had no significant influence on capillary absorption, regardless of the amount of cement replacement. Only in mixtures made with ash F6 the capillary absorption coefficient was significantly decreased. Like all fluid transport processes through concrete, the rate of sorption is governed by the pore system [28]. Therefore, the reduction in sorption rate can be related to the reduced porosity due to the addition of ash F6.

![Fig. 4. Capillary absorption coefficients of concrete](image)

4. Conclusions

The incorporation of WFA in concrete as a partial cement replacement can have a large impact on the fresh and hardened concrete properties. Mixtures made with WFA F6 with a particle size distribution close to that of cement exhibited increased cohesivity, which in turn reduced slump and required more energy for compaction of concrete. At the same time, this type of ash accelerated hydration and stiffness development. The increased cohesiveness of the fresh concrete resulted in concrete with a denser structure and lower porosity, which had a positive influence on the compressive strength and permeability. All these effects can be explained by the filler and filling mechanisms. Mixtures made with WFA F4 that had a similar chemical composition to ash F6 but consisted of coarser particles showed no evidence of the filler effect mechanisms. This suggests that governing mechanisms introduced by the cement replacement with fly ash are connected to the physical interactions between the phases.

The water demand of the concrete is balanced by the amount of water adsorbed by the WFA particles and the amount of released entrapped water due to the filling effect. The results obtained on mixtures containing F4 and F6 ashes indicate that there may be an optimum content of WFA at which the water demand could be reduced. Increasing the WFA content beyond the optimum will increase the quantity of water required to achieve targeted workability.

The research presented shows that replacing up to 45% of the cement with WFA has very little effect on capillary absorption and can give concrete with a sufficiently high compressive strength to be suitable for construction purposes.

Acknowledgements

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References


