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Case Report

Improving sustainability of mortar by wood-ash and Nano-SiO2

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A R T I C L E I N F O

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A B S T R A C T

Cement production charges the environment with CO_2 pollution and exhausts natural stocks of limestone and clay. Cement was partially replaced with Wood Ash (WA), and nano-SiO_2 (NS) was added to balance the strength loss. Mortar specimens were prepared by replacing the cement with WA up to 20 %, 30 %, 40 %, and 50 %, using constant-flow mortar. The flexural and compressive strength results indicated that although adding 40 % of WA reduced the mortar's strength, it still remained in an acceptable zone (higher than 20 MPa). The addition of 2.5 % NS recovered the strength loss caused by replacing cement with 40 % WA.

1. Introduction and research background

Cement production contributes to a significant extent to CO_2 pollution [1]. Average cement production is around 2800 million tons/year, and estimations show that it will increase to 4000 million tons/year by 2050 [2]. The growth in population and the demand for cement-based construction contribute significantly to climate change through energy consumption, depletion of raw materials [3], the generation of greenhouse gases and dust, and consumption of large quantities of raw materials [4,5]. To mitigate these adverse effects, researchers focus on replacing cement with waste materials to enhance the sustainability of cement mortar-based construction materials, given that producing one ton of cement consumes 1.5–1.75 tons of raw resources [6] and around 400 kg of greenhouse gases are emitted for every 600 kg of cement produced [7].

The cement makes up around 10–40 % of the total mortar mixture volume; it has a significant bond strength that links the particles of sand together while it reacts with the water [8]. The 2016 EU construction and demolition waste management protocol highlights the advantages of recycling that go beyond financial gains, employment generation, and less landfilling. Eethar, Alya'a, and Zinad (2020) replaced cement with wood ash (from charcoal) and found that the mechanical and physical properties of the mixture decreased [9]. Large stocks of WA are available and used for landfilling (70 %) [10] as wood is frequently burned for energy [7]. In Great Britain an estimated amount of 2.8–8.9 kilotons of WA was generated in 2017, and this production is expected to increase to 15.7 kilotons by 2050 [11]. In Austria, the total amount of WA produced in heating plants in 1995–96 was about 68000 tons per year, and increased in 10 years to 104000 tons per year [12]. Ash from wood is more environmentally friendly [13] than fly ash and offers flexibility in formulation, as its density, specific gravity, composition, etc. [14] and quality vary with wood species [15]. Adding nanomaterial to fly ash made it possible to achieve new properties of mortar [16]. The reaction of the pozzolanic materials (WA, NS) with calcium hydroxide increases the amount of C–S–H chemical bonds, resulting in highly dense concrete [17]. This density adds more strength and durability to the material. Wood ash results from a much more environmentally friendly process than fly ash. The chemical composition of wood ash differs from that of fly ash, and it was investigated in the current study whether it is possible to achieve better physical and mechanical properties of the mortar (formulated with wood ash) and mixed with NS than in the case of fly ash. The current study examined the influence of partly replacing cement with wood ash and adding NS to reinforce the strength loss.

2. Materials

The conventional mortar M0 was considered a reference mix. M2, M3, M4, and M5 were mixed with different amounts of WA to replace cement (Fig. 3).

2.1. Ordinary Portland cement (OPC)

Produced by the Badoosd factory as shown in Tables 1 and 2.

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Table 1
Chemical composition of the OPC.

Property	Test result (%)	Standard IQS, NO.5/1984 (%)
1. Oxide composition:		
Alumina, Al_2O_3	4.7	3–8
Silica, SiO_2	20.1	17–25
Ferric Oxide, Fe_2O_3	3.5	0.5–6
Lime, CaO	63.7	60–67
Sulphuric Anhydride, SO_3	3.1	Max. 2.8
Magnesia, MgO	0.7	Max. 5
2. Compound composition:		
C_3A	7	
C_2S	18	
C_3S	53	

Notation: C=CaO, S=SiO_2, A = Al_2O_3, these results are provided by the manufacturer.

Table 2
Physical properties of the OPC.

Property	Test result	Standard IQS, NO.5/1984
Fineness % (Residue on sieve size 0.15 mm)	3	Max. 10
Specific surface "Blaine" (cm^2/g.)	3447.3	Min. 2250
Initial setting time (min.)	142	45
Final setting time (min.)	384	max 600
Specific gravity	3.23	–
Compressive strength (MPa)		
At 3-day	20.12	min 16.0
At 7-day	26.98	min 24.0

The manufacturer provides these results.

2.2. Aggregate

A natural aggregate with properties and particle sizes according to ASTM C136 was used after being tested by OSZ at NTU.

2.3. Water

Tap water, according to ASTM C1602, was used.

2.4. Wood Ash (WA)

It was obtained from open-field coal burning at an average temperature of 200°C. It was dried and carefully homogenized. Table 3 Provides the physical and chemical properties of the WA, as tested by OSZ at NTU and TU (Fig. 1).

Table 3
Chemical and physical properties of WA.

Chemical Properties					
Wt. %	%	Wt. %	%	Wt. %	%
Na_2O	2.58	**CaO**	6.08	**As_2O_3**	–
MgO	5.88	**TiO_2**	0.15	**SrO**	0.49
Al_2O_3	1.16	**Cr_2O_3**	–	**ZrO_2**	–
SiO_2	52.03	**MnO**	0.05	**BaO**	0.16
P_2O_5	3.54	**Fe_2O_3**	1.00	**PbO**	–
SO_3	1.46	**NiO**	–	**CszO**	0.01
Cl_2	0.98	**CuO**	–	**Rb_2O**	–
K_2O	8.57	**ZnO**	0.06	**Loss of ignition (LOI)**	6.10
Sum					**90.30**

Physical properties		
Specific gravity	Mean size (μm)	Density (kg/m^3)
2.39	0.1	715

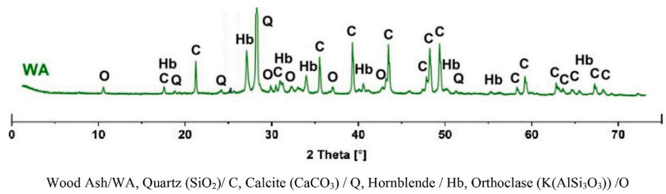

Fig. 1. XRd. of WA taken at TUM (Laboratory of the Technical University of Munich, Germany).

2.5. Nano silica (NS)

Serves as a desiccant, thus decreasing the void ratio, filling the voids, and supporting the creation of newer C–S–H chemical bonds. It is a highly reactive pozzolanic material, a 99.97 % SiO_2 with a particle size of 14 nm, fineness of 190 m^2/g, with a 0,67 % loss on ignition at 1000°C, density of 13.5 kg/m^3, and 0,9 % retain on a 45 μm sieve. Protective clothing was used during sample preparation, considering the precautionary recommendations associated with nanoparticles [18].

2.6. Superplasticizer (HRWRA)

It was provided by SIKA company and conformed with ASTM C494 (A, F, and G) with a density of 1.208 kg/l, chloride content > 0.1, alkaline content > 5.

2.6.1. Method

Seven mix types of mortar, M0 (control), M1, M2, M3, M4, M5, M6 were prepared with W/C as given in Table 4. Cubic molds of 50x50 × 50 mm dimension were used for casting mortar specimens for the compressive strength tests. Prism molds of 40x40 × 160 mm dimension were used for casting mortar specimens for the flexural strength, density, and water absorption tests. After casting, all the test specimens were stored at room temperature, de-molded after 24 h, and placed into a water-curing tank at 24–34 °C according to ASTM C192 until testing. While replacing cement with 20, -30, −40, and 50 % WA, the flow rate was kept fixed parallel with different water content. After testing the first six mixes (from M0 to M5), mix M4 (based on its high strength values relative to the cement replacement ratio) (Fig. 2) was selected to be additioned with NS and became mix M6, as shown in Table 4. The fresh concrete was tested for workability by flow table (produced by UTEST company), matching the specifications of ASTM C230. The amount of water content in the mortar was selected to keep a 115 % flow rate, according to ASTM C1437.

After sample preparation, compressive and flexural strength, dry density, water absorption, and void ratio tests were performed on hardened concrete samples. **Compressive strength (Cs) was** tested according to ASTM C109/109 M, **Flexural strength (Fs)** was tested according to ASTM C348 by MATEST machine, one-point loaded in the middle, on the upper surface, with 100-mm space between the two fixed supports. **Dry density (γ_d), Water absorption (Wb), and Void ratio (Vr)** tests were performed according to ASTM C642 (see Fig. 4).

3. Results and discussion

Compressive and flexural strength for 7 and 28-day, density, void ratio, and water absorption are shown in Table 5. The image of the microstructure for WA and mortar with WA and NS is shown in Fig. 5.

3.1. Discussion

C_3S is responsible for most of the early strength (first 7-day) as

$$2Ca_3SiO_5 + 7H_2O \rightarrow 3CaO.2SiO_2.4H_2O + 3Ca(OH)_2 + 180\ kJ$$

C_2S also affects the strength of concrete but in a much more slowly

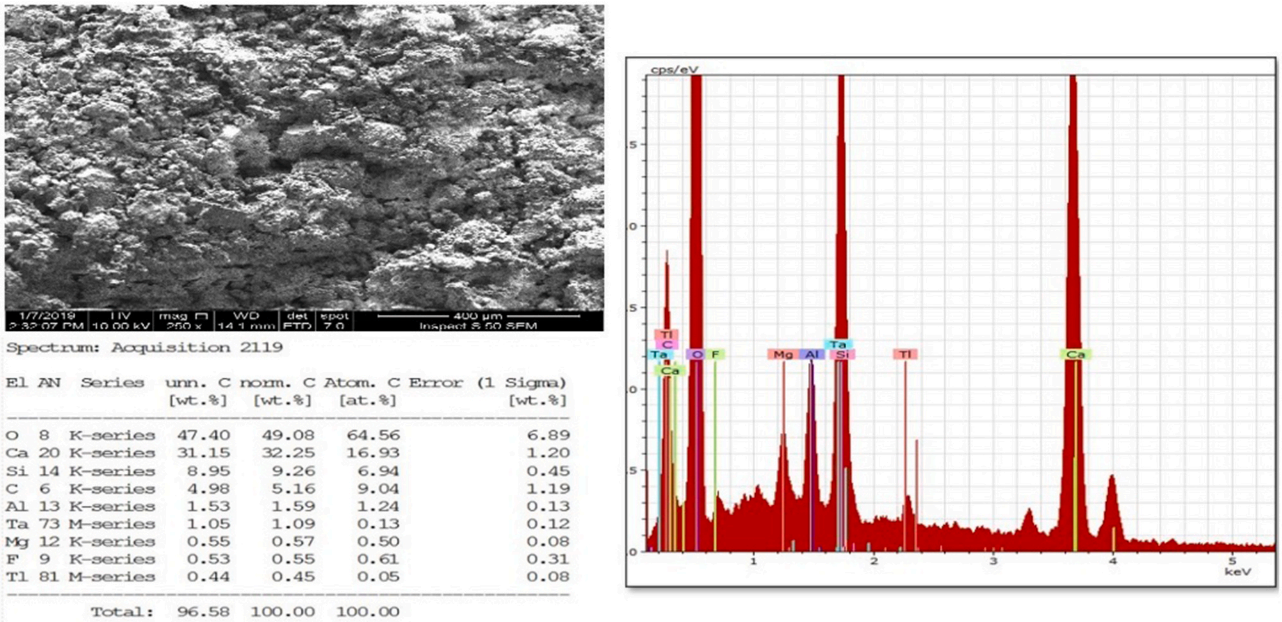

Fig. 2. SEM & XRd. Of M4 at 28-day taken at UT (Laboratory of the University of Technology, Iraq, Baghdad).

Table 4
The 7-mix types based on cement replacement proportions.

Mix	Cement (kg/m^3)	Sand (kg/m^3)	W/C	HRWRA (%)	WA (kg/m^3)	NS (kg/m^3)
M0	540	1485	0.48	0	0	0
M1	540	1485	0.32	0.4	0	0
M2	432	1485	0.35	0.4	108	0
M3	378	1485	0.38	0.4	162	0
M4	324	1485	0.41	0.4	216	0
M5	270	1485	0.45	0.4	270	0
M6	324	1485	0.45	0.4	216	13.5

Table 5
Compressive strength results.

Mix	Cs 7-day (MPa)	Cs 28-day (MPa)	Fs 7-day (MPa)	Fs 28-day (MPa)	Ɣd kg/m^3	Vr %	Wb
M0	24.8	32.48	6.4	7.21	2318	5.32	4.35
M1	28.17	35.86	7.1	7.6	2380	4.85	4.18
M2	21.31	32.17	5.7	6.23	2285	4.51	4.35
M3	18.41	30.27	5.2	5.6	2215	4.22	4.78
M4	16.81	28.54	4.9	5.3	2175	3.82	5.2
M5	13.67	21.12	4.1	4.7	2062	3.15	5.9
M6	23.21	31.84	6.3	6.91	2230	2.22	4.26

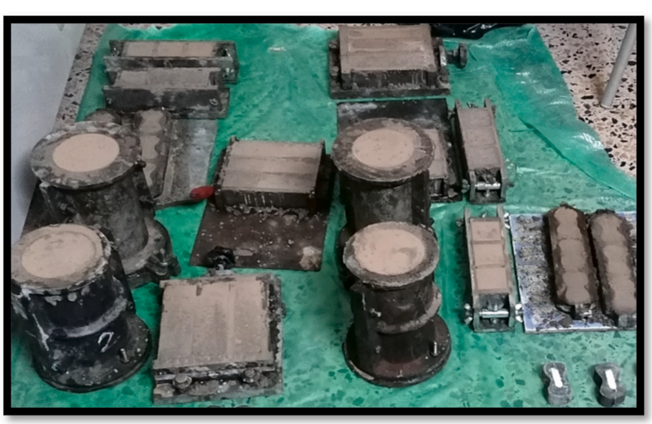

Fig. 3. Casting molds and specimens (Photo: Zinad).

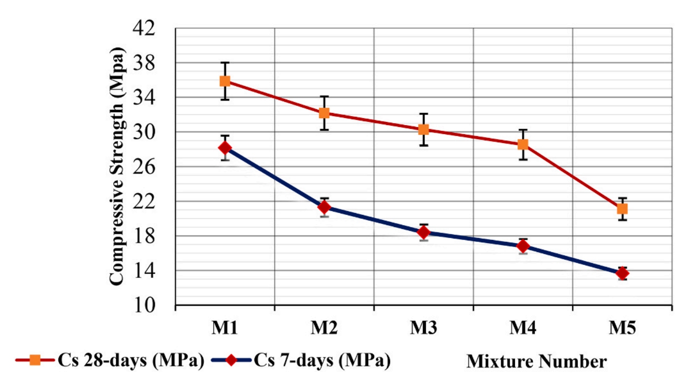

Fig. 4. Cs at 7 and 28-day with 0,- 20,- 30,- 40,- and 50 % WA

process of hydration:

$$2Ca_2SiO_4 + 5H_2O \rightarrow 3CaO.2SiO_2.4H_2O + Ca(OH)_2 + 58.6\ kJ$$

Hardened cement paste ettringite occupies about 15–20 % by volume, calcium silicate, C–S–H occupies about 50–60 %, calcium hydroxide (lime) occupies 20–25 %, and voids occupy about 5–6 % [19]. Supplementary cementitious materials improve mortar strength by promoting the formation of new C–S–H bonds through reactions with Ca $(OH)_2$. From the three formulations C_2S, C_3A, and C_3S, it was stated earlier that C_3S can be expected to have the highest Strength Activity Index (SAI) after 7-day casting; our results support this statement and also conform with Salih et al. (2020) [20]. Adding 0.4 %, HRWR increased compressive and flexural strength while reduced the mortar's water content and void ratio. HRWR forms long polymer chains during hydration, blocking water from entering the voids and enhancing strength. M1, without WA but with a superplasticizer, showed promising results compared to M0. Adding 0.4 % HRWR to the mixture increased compressive and flexural strength by 9 % and 5 % compared to the control mix (M0) after 28 days. By replacing 20 % of cement with WA,

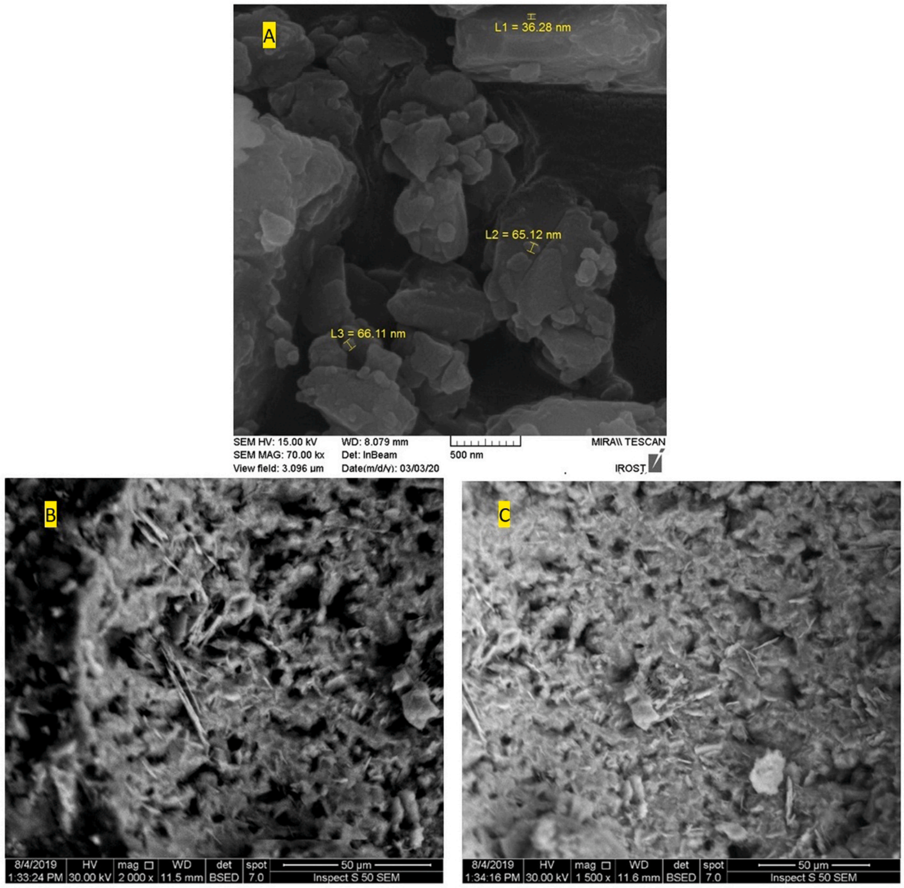

Fig. 5. **A**. SEM micrograph for WA and nano-SiO, **B.**SEM micrograph for ordinary cement mortar, **C.** SEM micrograph for cement mortar with WA and NS (Image: OSZ).

the mortar's compressive and flexural strength decreased by 10 % and 18 %, respectively (M2 to M1). Replacing 30 % of cement, the upper strengths decreased by 16 % and 26 % (M3 to M1). The decrease in compressive strength when comparing mixture M4 and M5 with M1 was 20 % and 40 %, respectively, while flexural strength was 30 % and 38 % after 28 days. Comparing mixture M6 with M4, the compressive and flexural strength increased by 12 % and 28 % due to the addition of NS.

The density and void ratio decreased by replacing cement with WA because the WA density was lower than the density of cement.

The IBC and ACI 318 Standards require a minimum Cs of 17.3 MPa for structural concrete. All seven mortar mixtures exhibited higher Cs at 28 days than this minimum requirement. However, mixture M5, with 50 % WA, had the lowest Cs: 21.12 MPa. Although this still exceeds the minimum requirement, M5 is not considered optimal because of a significant decrease in strength compared to the amount of WA added. M4 can be promoted as optimal because its favourable ratio of Cs decreases relative to the increase of WA content, having a 28,54 MPa Cs, with acceptance FS, VR, WB, and Ɣd. On the other hand, 40 % of WA to replace cement is an acceptable percentage to achieve sustainability, as it significantly contributes to decreasing cement consumption by using waste materials like WA.

The growth in strength for mixture M6 is due to NS, which enters mortar voids and fills them, resulting in a lower void ratio and, thus, higher strength. The reason is the high surface area of the nanomaterial, which works as a nucleation site for the precipitation of C–S–H gel, enlarging the relative surface area to react with calcium hydroxide and form further C–S–H bonds. The NS can react with $Ca(OH)_2$ crystals, thus making the Interfacial Transition Zone (ITZ) and binding cement paste denser, as shown in Fig. 5. Mixing NS reduces cement pastes' calcium leaching rate and increases their durability [21]. The high percentage of WA replacing cement makes cement mortar have a lower density than conventional mortar, which is advantageous. Mortar with WA and NS showed reduced water absorption due to changes in pore structure caused by their reaction with $Ca(OH)_2$ during OPC hydration. This resulted in decreased voids, lower water absorption, and reduced density, as indicated in Fig. 5. Fig. 5B shows that the WA and NS particles filled the voids and formed an ITZ with new C–S–H bonds, which make the cement mortar denser, compared to Fig. 5B.

4. Conclusions

This article suggests a more sustainable construction practice by substituting up to 40 % of the environmentally weak cement with WA and reducing the raw material need and the cost of cement mortar. Thus, the principle of more sustainable green construction is achieved.

Replacing cement with WA has double environmental advantages: a waste material gets used, and cement production is lowered. The major question was if the mechanical properties of WA cement mortar meet the expected strength after replacing cement with WA. Following the tests, only one mixture was considered optimal, the one with 40 % WA, but even in this case the considerable amount of WA addition resulted a 17.3 % and 25.5 % decrease in compressive and flexural strength. Although these strength values are far above the expected minimum, they are lower than the values of the conventional mortar. In order to improve the SAI of the mortar containing 40 % WA, in continuation samples were supplemented with NS. The addition of NS fulfilled the expectations, resulting in enhanced SAI, reduced void ratio, and reduced water absorption of the 40 % WA + NS samples. As consequence, the amount of raw material need (like limestone, etc.), the CO_2 pollution and the landfills caused by cement production can all be reduced.

Replacing cement with 40 % WA and adding NS made it possible to produce a more environmentally friendly and sustainable cement mortar with mechanical properties that are equally good to those of conventional cement mortars.

CRediT authorship contribution statement

Omar Saber Zinad: Writing – original draft, Methodology, Formal analysis, Data curation, Conceptualization. **Csilla Csiha:** Writing – review & editing, Validation, Supervision, Project administration, Formal analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data is made available in the text

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