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Application of carbon black in conductive fly ash geopolymer mortars

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Abstract. Materials with enhanced electrical properties are widely studied concerning their future application in smart or self-sensing constructions. Carbon black is known as one of the inexpensive multipurpose admixtures used in polymeric materials. This study is focused on the application of carbon black in fly ash geopolymer mortar to enhance its conductivity and other electrical properties. Geopolymer samples were prepared with the carbon black content from 0.5 to 4.0% to evaluate their performance in selected electrical properties (conductivity, resistance, capacitance). Further investigations included its influence on the mechanical properties (compressive and flexural strength) and microstructure of the binder (mercury intrusion porosimetry, SEM). Despite the considerable improvement in conductivity, the amount of conductive filler over 2% was also associated with increased porosity and reduced compressive strengths.

1. Introduction

Carbon black is an amorphous form of carbon produced by partial combustion or thermal decomposition of hydrocarbon materials. Since it consists of elementary carbon particles of high specific surface, it is widely used in many industrial applications as a pigment or light weight filler with high chemical and thermal stability [1]. Apart from the major application for rubber and plastics reinforcing, it is also incorporated in conductive composites due to its permanent conductivity properties and affordable price [2].

Structural composites with enhanced conductivity can be used in smart structures using their selfsensing properties, i.e. capability to sense strain, internal damages etc. based on the evaluation of composite bulk electrical conductivity/resistivity variation that resulted from the external condition changes. Smart structure is thus designed as a sensor itself and it is suitable to provide easy and cost effective structural health monitoring that does not require installation of embedded or additional sensors [3]. This approach is adopted for number of advanced applications for traffic monitoring solutions (vehicular speed, weighing in motion, traffic density) [4] or thermal and humidity control in structure related to its self-heating properties. For instance, Mingqing [5] successfully incorporated carbon black into mortar for indoor electrical floor heating slab and similar concept is applied in case of possible deicing solutions for large airport areas or car parks [6]. Moreover, the introduction of carbon conductive admixture in cement matrix increases its electromagnetic shielding ability. Electromagnetic interference shielding of rooms in buildings is needed for protecting electronics, especially those associated with strategic systems such as aircraft, nuclear reactors, transformers, communication and control systems, etc. [7].

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI. Published under licence by IOP Publishing Ltd 1 The use of carbon black or other conductive fillers has been investigated predominantly in polymeric and cement matrices but this study deals with their use in a fly ash-based geopolymer matrix. Geopolymers are a group of inorganic mineral binders derived from the reactions of aluminosilicaterich precursor under high alkaline conditions. Their polymeric and highly coordinated three-dimensional structure formed by SiO₄ and AlO₄ units was reported by prof. Davidovits who firstly described the geopolymerization mechanism [8]. Convenient source materials for geopolymer synthesis include any natural or manufactured materials with high Si and Al content in amorphous form, namely low Ca fly ash, metakaolin and calcined kaolin or other clays [9] that are mixed with alkali hydroxide/alkali silicate activator solution [9, 10]. Recently, geopolymers has attracted considerable attention because of their early compressive strength, good chemical resistance and excellent fire resistance behaviour [11]. Depending on the raw materials chosen and curing conditions, alkali activated binders can represent more environment friendly alternative to the ordinary Portland cement [12].

Studies discussing the role of carbon black in geopolymer binders are limited, authors rather used other carbon conductive fillers; graphite, carbon fibres (CF), carbon nanotubes (CNTs) or graphene. Electrical conductivity of fly ash geopolymer binder as such is affected by the activator solution, liquid activator/ash ratio (L/A) and frequency spectrum [13]. The improvement of both electrical and mechanical performance of fly ash geopolymer doped with CNTs was studied by Saafi et al. [14] who states that the optimal concentration is 0.5%. Mackenzie et al. [15] prepared clay-based geopolymers with 0.26% of single wall carbon nanotubes (SWCNTs) and the same concentration of graphite for comparison. As is the case with CNTs, CF is effective conductive filler with the percolation threshold at 0.5% concentration [16]. In another study, partial replacement of CF by carbon black in cement-based matrix maintained the conductivity while lowering the costs and increasing the workability [17]. Excellent results were achieved with application of graphene in conductive composites. Only 0.35% addition resulted in 209% increase of conductivity and lower porosity accompanied by significant increase of strength and modulus of elasticity [18].

2. Materials and methods

Fly ash (FA) from the black coal combustion with chemical composition given in Table 1 was mixed with commercially sold sodium silicate solution (molar ratio $SiO_2/Na_2O = 1.6$, chemical composition in table 2) to prepare the geopolymer binder.

	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	S _{total}	Na ₂ O	K ₂ O
(%)	49.82	24.67	7.50	3.91	2.68	0.91	0.70	2.78

 Table 1. Chemical composition of fly ash.

Table 2. Chemical composition of water glass solution.

_	SiO_2	Na ₂ O	H_2O
(%)	26.43	16.61	56.96

Conductive filler - carbon black Vulcan 7H was added in concentration of 0.5 to 4.0% together with a small amount of water and the dispersing agent, Triton X-100 (2% solution). After proper homogenization, standardized quartz sand with a maximum grain size of 2.5 mm was added as an aggregate together with remaining amount of water. Total amount of water in each mixture varied in order to reach the same workability of the fresh mortars (mortar diameter 180 ± 10 mm using the flow table method, EN 1015-3 1999). Mix compositions are shown in table 3.

	REF	S 0.5	S 1.0	S 2.0	S 3.0	S 4.0
Fly ash (g)	350	350	350	350	350	350
Sodium silicate (g)	280	280	280	280	280	280
Carbon black (g)	-	1.75	3.5	7	10.5	14
Sand (g)	1050	1050	1050	1050	1050	1050
Triton X-100 2% (ml)	-	1.75	3.5	7	10.5	14
Water (g)	35	35	35	40	45	50

 Table 3. Mix design of fly ash geopolymer mortars.

Fresh mortars were placed into prismatic moulds ($40 \times 40 \times 160$ mm) and covered with plastic sealant to protect the binder from moisture loss. After 2 hours in laboratory conditions ($22 \pm 2^{\circ}$ C, $\varphi = 45 \pm 5\%$), the samples were thermally treated at 40°C for 24 h. Demoulded specimens were stored in laboratory till the age of testing.

Before the measurements, the prepared prismatic samples were dried at 105°C to reduce the influence of moisture on assessed properties. They were characterized by impedance spectroscopy in the range of 40 Hz to 1 MHz using an Agilent 33220A sinusoidal signal generator and an Agilent 54645A dualchannel oscilloscope. In order to perform impedance analysis, the prismatic specimens were placed between parallel brass electrodes (30×100 mm) so that a distance between electrodes was 40 mm. The output voltage of the signal generator was 5.5 V. The input values for the electrical capacity and the resistance of the oscilloscope were 13 pF and 1 M Ω , respectively. These instruments were assembled for fully automated measurement. At higher range of frequencies, 100 MHz up to 3 GHz, R&S ZNC vector analyzer with DAK 12 coaxial probe manufactured by Speag has been used. In this frequency spectrum the electrical conductivity and relative permittivity as a function of frequency have been measured.

Geopolymers were tested for their mechanical properties (compressive and flexural strength). Micromeritics Poresizer 9310 and scanning electron microscope (SEM) Tescan MIRA3 XMU were used to assess the microstructure and porosity of the geopolymers.

3. Results and discussion

3.1. Electrical properties

Electrical resistance of all geopolymers decreased with the carbon black content and with higher frequency applied, as can be seen from Figure 1. At low frequency, samples with carbon black content 2% and higher exhibited considerably lower resistance, initial resistance of the reference sample dropped from 21.3 to less than 3 M Ω . As to frequency from 2 kHz, minor differences among all samples and slower decrease in resistance were observed.

Regarding the influence of frequency, similar trends were noted at changes of electrical capacitance (Figure 2) but with the inverse effect of the filler content. Capacitance of geopolymers S2.0 to S4.0 up to 200 Hz fluctuated and the curves overlapped. At 100 Hz, the electrical capacitance of these geopolymers was twice as high as of the reference sample with the capacitance 55.9 pF. Above 1000 Hz, the curves were smooth and sample S4.0 achieved significantly higher capacitance.



Figure 1. Variation in electrical resistance of FA geopolymer mortars with different carbon black content.



Figure 2. Variation in electrical capacitance of FA geopolymer mortars with different carbon black content.

As expected, higher carbon black content in geopolymers led to improvement in electrical conductivity properties across the frequency spectrum shown in Figure 3. In fact, the resistance of samples S3.0 and S4.0 was nearly the same but S4.0 was characterized by increased capacitance from frequency 1000 Hz and enhanced conductivity at all frequencies. Unlike resistance and capacitance, the electrical conductivity increases at higher frequency.



Figure 3. Electrical conductivity of FA geopolymer mortars with different carbon black content at chosen frequencies.

3.2. Mechanical properties

Variations in mechanical performance of geopolymers depending on the carbon black content are displayed in Figure 4. The flexural strength was not negatively affected by the conductive filler and remained constant around 5 MPa. On the other hand, the compressive strength gradually decreased and at carbon black content of 2–4% attained approximately 50% strength in comparison with the reference sample.



Figure 4. Mechanical properties of FA geopolymer mortars with different carbon black content.

3.3. Microstructure

Excessive amount of water needed during the preparation of samples with higher carbon black content caused an increment in total porosity of the binders (see Figure 5). At low carbon black concentrations, the cumulative intruded volume was about 0.080 cm³·g⁻¹ but it increased to more than 0.095 cm³·g⁻¹ at composites with 2–4% carbon black concentration. This fact is in accordance with reduced compressive strength.

SEM imaging of the composites did not reveal the real distribution of carbon black particles within the matrix because of their exceptionally small dimensions (figure 6, picture A). Agglomerates of colloidal size carbon black particles are completely separated during the mixing process, hence no difference in microstructure of REF and S4.0 samples (pictures B, C) is clearly visible. In this case, electron microscopy was not a convenient method to evaluate the quality of their dispergation.



Figure 5. Cummulative intruded volume of FA geopolymer mortars with different carbon black content.



Figure 6. SEM images, A: carbon black particles (scale: 500 nm), B: sample REF (scale: 10 μm), C: sample S4.0, (scale: 10 μm).

4. Conclusions

This study was performed to evaluate the electrical properties of fly ash geopolymer mortars with carbon black conductive admixture. Geopolymers with carbon black concentration of 0.5–4.0% were analysed by impedance spectroscopy to assess their electrical conductivity, resistance and capacitance, these experiments were followed by testing of the mechanical performance. Carbon black content from 2% resulted in reduced electrical resistance and enhanced capacitance of the mortars, especially at low frequencies up to 100 Hz. At higher frequency range, both resistance and capacitance of all samples showed minor differences. Further increase of carbon black to 4% content resulted in a considerable improvement of electrical conductivity and slight increase in capacitance. Although the nanoparticles of carbon black effectively enhanced the electrical properties, their incorporation into binder structure was accompanied by additional water demand and increased pore volume of hardened specimens determined by the mercury intrusion porosimetry. This led to the deterioration of the compressive strength but the flexural strength of all samples was stable and not negatively affected. As SEM imaging confirmed complete dispergation of carbon particles, carbon black represents a low-cost and effective nanomaterial for conductive composites. Possible combination with another conductive admixture (for instance carbon fibers) may help to eliminate some negative aspects of their application.

Acknowledgements

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