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Mechanical properties and ablation behaviour of nuclear sacrificial materials containing graphene sulfonate nanosheets

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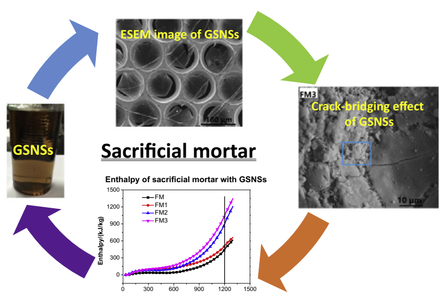
HIGHLIGHTS

- Mechanical properties and ablation behaviour of sacrificial materials are studied.
- Optimal amount of GSNSs is 0.1 wt% in sacrificial materials.
- Flexural strength of sacrificial mortar increases by 16.22% via GSNSs.
- Compressive strength of sacrificial mortar is up by 24.44% via GSNSs.
- Ablation rate of sacrificial mortar decreases by 50.33% via GSNSs.

GRAPHICAL ABSTRACT



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ABSTRACT

Graphene and its derivatives have been attracting the widespread interest of researchers because of their capability to improve several properties (e.g. mechanical properties, durability, anti-corrosion) of cementitious composites. This paper presents an experimental study on the influence of graphene sulfonate nanosheets (GSNSs) on mechanical properties and ablation behaviour of ferro-siliceous sacrificial cement paste and mortar, including flexural strength, compressive strength, microstructure, porosity, and thermal properties. Based on the results of differential scanning calorimetry and decomposition temperatures, the decomposition enthalpy of ferro-siliceous sacrificial cement paste and mortar was determined. It was found that, (1) the flexural strength and compressive strength of ferro-siliceous sacrificial mortar were increased by 16.22% and 24.44% respectively with the addition of 0.1 wt% GSNSs; (2) the decomposition enthalpy of ferro-siliceous sacrificial mortar was increased by 14.65%, 101.33%, and 135.15%, when adding 0.03 wt%, 0.1 wt%, and 0.3 wt% GSNSs, respectively; (3) the optimum GSNSs content was 0.1 wt% considering the mechanical strength, microstructure, and ablation rate of ferro-siliceous sacrificial cement paste and mortar. These findings can guide the design of ferro-siliceous sacrificial composites, e.g., cement paste, mortar, and even concrete containing GSNSs.

1. Introduction

At present, cement and concrete composites are the most extensively building and construction materials. However, cementitious composites are quasi-brittle materials and inclined to crack since they have high compressive strength but relatively low flexural strength, and poor strain capacity [1]. Therefore, how to

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prevent cracking and improve the tensile strength are the main concerns in the field of cementitious composites [2–4]. Recent progress in nanotechnology provides opportunities to tremendously enhance the performance of cement or concrete composites through nano-sized particles or fibres, such as nano-SiO_2 [5–9], carbon nanotubes [10–15], carbon nano-fibre [15,16], nano-$CaCO_3$ [17], and nano-TiO_2 [18–20]. Nano-sized materials in cementitious composites are more effective than conventional reinforcements that are usually at millimetre scale, because they can control the formation and development of nano-size cracks at the initial stage [21]. The nanomaterials used in modified cementitious composites can be classified into zero-dimensional (0D) particles (e.g., nano-SiO_2, nano-TiO_2 and nano-$CaCO_3$), one-dimensional (1D) fibres (e.g., carbon nanotubes and carbon nano-fibre), and two-dimensional (2D) sheets according to the shape of nanomaterials.

Graphene and its derivatives, as new 2D nanomaterials, have drawn the most attention in science and engineering in recent years. Graphene has excellent electrical, optical, mechanical, and thermal properties. The average Young's modulus and tensile strength of grapheme are 1100 GPa and125 GPa, respectively [22,23]. The thermal conductivity of suspended graphene is 5300 W/mK [24]. The specific surface area of graphene can theoretically reach 2.63×10^6 m^2/kg [25], which offers more space for physical and chemical interactions between graphene and matrix material. Nevertheless, high production cost and difficulty in dispersing are the major obstacles that limit the application of graphene. As new kinds of nano-sized carbon materials, graphene derivatives (e.g. graphene nanosheets and graphene oxide nanosheets) also contain graphene sheets [26–34], both of which display a 2D sheet-like structure, and the thickness of them is generally less than 10 nm, that is, still at nano scale. Graphene oxide nanosheets (GONSs) are oxides of graphene nanosheets (GNSs), and thus the GONSs have oxygen functional groups that scattered in the 2D sheet-like structure of GONSs. The interaction force between GONSs, namely, van der Waals force can be significantly modified due to the oxygen functional groups, hence the dispersive capacity of GONSs in water can be improved [35]. Shamsaei et al. [36] has summarized the excellent performance of graphene and graphene-based nanosheets, and they point out that graphene and graphene-based nanosheets possess extraordinary mencanical, chemical, thermal and electrical properties, enabling attractive applications, ranging from structural strength/durability improvement, anti-corrosion, to self-cleaning surfaces and energy saving. In addition, the GNSs and GONSs are cheaper to produce than that of graphene.

So far, much research has been done on the effects of GNSs and GONSs on properties of cementitious composites. Ranjbar et al. [37] found that the compressive strength of geopolymer increased by 44%, and that the flexural strength of geopolymer was raised by 116%, respectively, if 1% (mass fraction) GNSs were added. Sun et al. [38] reported that both the flexural and compressive strengths of oil well cement paste were improved because of the addition of GNSs. Du et al. [39] pointed out that the chloride diffusion of concrete was reduced by 80%, when 1.5% of graphene nanoplatelet was added. Mokhtar et al. [40] found that the tensile strength of cementitious composites was enhanced by about 41% due to the addition of 0.03% GONSs. Lv et al. [41] reported that the tensile, flexural and compressive strengths of cementitious composite were increased by 78.6%, 60.7% and 38.9%, respectively, when 0.03 wt% (with respect to weight of cement) GONSs were added. The previous research also indicated that GONSs could regulate the cement hydration process, and further improve the microstructure of cementitious composite [42]. GNSs and GONSs could bring certain self-sensing capability to cementitious composites [30], which was similar to the function of carbon fibre [43]. In addition, Sha et al. [44,45] suggested that understanding the failure behaviour and failure mechanism of graphene is also important so as to promote its structural and functional applications.

The safety problem of nuclear power is becoming increasingly acute on a global scale. Sacrificial concrete is the hotspot but also a difficulty in the modern nuclear technology. Sacrificial concrete, as a key component of core catcher in nuclear power plant, is designed to prevent leakage of radioactive materials from reactor containment in severe nuclear accident [46]. The corium temperature can be reduced via the interaction between sacrificial concrete and corium. In addition, the physical and chemical properties of the corium can also be modified. Accordingly, the reliability of the core catcher can be improved. As one kind of sacrificial concrete, ferro-siliceous sacrificial concrete is mainly used in the third generation nuclear technology that is the most advanced technique at present. Thermal performance of ferro-siliceous sacrificial concrete, particularly its ablation (erosion of material due to high temperature) behaviour plays a critical role in severe nuclear accident mitigation. Therefore, the ablation behaviour of ferro-siliceous sacrificial concrete should be studied urgently due to the increasing application of ferro-siliceous sacrificial concrete in nuclear power plant. Although graphene and its derivatives have been extensively investigated currently, there is still no available information concerning the effects of graphene sulfonate nanosheets (GSNSs) on microstructure, mechanical properties and ablation behaviour of cementitious composites. The $-SO_3H$ contained in GSNSs plays a similar role of $-OH$ in GONSs, so the GSNSs may also find their applications in cementitious composites [47]. Furthermore, the total cost of production for GSNSs is about 11% lower than that of GONSs.

The main purpose of the work is to experimentally study the effects of GSNSs on mechanical properties and ablation behaviour of ferro-siliceous sacrificial cement paste and mortar. The flexural strength, compressive strength, thermal analysis, microstructure, porosity, and thermal conductivity of ferro-siliceous sacrificial cement paste and mortar with different contents of GSNSs were comprehensively explored. Based on the experimental results of differential scanning calorimetry and decomposition temperatures, the decomposition enthalpy of ferro-siliceous sacrificial cement paste and mortar was determined. Finally, the ablation behaviour of ferro-siliceous sacrificial cement paste and mortar was investigated in a quantitative manner.

2. Experimental program

2.1. Materials

P•II 52.5 cement, silica fume, and fly ash (Class I) were utilized in the paper. The chemical composition and physical properties of

Table 1
Chemical composition and physical properties of cement, silica fume, and fly ash.

Materials	Cement	Silica fume	Fly ash
Chemical composition	(wt.%)		
CaO	64.70	0.77	8.38
SiO_2	20.40	96.18	47.96
Al_2O_3	4.70	0.96	30.46
Fe_2O_3	3.38	0.85	5.91
MgO	0.87	0.74	2.60
SO_3	1.88	0.50	1.32
K_2O	0.83		1.61
Na_2O			1.76
Loss	3.24		
Physical properties			
Specific gravity	3.15	2.22	
Specific surface (m^2/kg)	362.20	2.79×10^4	
28d Compressive strength (MPa)	62.8		