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Research paper

# Molecular dynamics study on the weakening effect of moisture content on graphene oxide reinforced cement composite

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## HIGHLIGHTS

- Water in GO/C–S–H interlayer weakens the interaction between $Ca_{C-S-H}$ and $O_{GO}$.
- Water in GO/C–S–H interlayer weakens structural H-bond.
- The connection between interlayer Ca and $COO^-$ group is the most stable.
- Tensile strength of GO/C–S–H composite is reduced with moisture content increasing.

## ABSTRACT

Molecular dynamics is utilized to investigate the effect of moisture content on the molecular structure and bonding in the interlayer region between calcium silicate hydrate (C–S–H) and graphene oxide sheet functionalized by deprotonated carboxyl (COO), hydroxyl (C–OH), epoxy (C–O–C) group. The interfacial bonding is dependent on $Ca-O_{COO}$, $Ca-O_{C-OH}$, and $Ca-O_{C-O-C}$ bond as well as H-bond. These interlayered chemical bonds are sensitive to interlayer water and both of them can be degraded by moisture content increasing, resulting in a decrease in failure strength of GO/C–S–H composite. Among these interlayer chemical bonds, $Ca-O_{COO}$ bond possesses the highest strength and stability.

## 1. Introduction

With large specific surface area, hydrophilic nature, and outstanding mechanical properties [1–4], graphene oxide (GO) has attracted significant interests as a reinforced material for cement-based materials [5–10]. It has been reported that through the incorporation of GO into the cement-based materials, both tensile strength and flexural strength can increase up to 50%.

Based on the previous experimental results, the reinforcement of GO for cement-based materials is mainly attributed to the following two aspects. Firstly, the incorporation of GO nanosheets can regulate the microstructure of binder phase in cement system by promoting hydration and changing the crystalline forms of hydration products [11]. Secondly, the GO nanosheets can fill the defective region in the cement paste to densify its pores structure [12]. As for above second aspect, Hou and his co-workers [13,14] investigate the bonding performance and interacted mechanism between GO sheets and C–S–H matrix on the molecular level by means of computation. It was found that the improvement of the tensile strength and toughness of the cement materials mixed with GO sheets is mainly attributed to the bonding effect of GO sheets distributed in defective region of C–S–H gel (the main products in cement-based materials). The bonding effect is significantly determined by the functional groups rooted on the GO sheets [15–20]. Interestingly, it has been reported that graphene nanopores or membranes functionalized by different functional groups exhibits different enantioselective molecular transport properties [21,22].

It has been reported that the structure and mechanical property of GO are significantly influenced by environmental humidity [23,24]. For GO reinforced cement composite, water molecules are always concentrated in the interlayer void between GO and hydrates even after drying as a result of the hydrophilic nature of GO. The reinforced effect of GO for cement materials may be, to some extent, influenced by the penetrated water. However, there is only few study in this field.

In this paper, molecular dynamics (MD) is utilized to construct the

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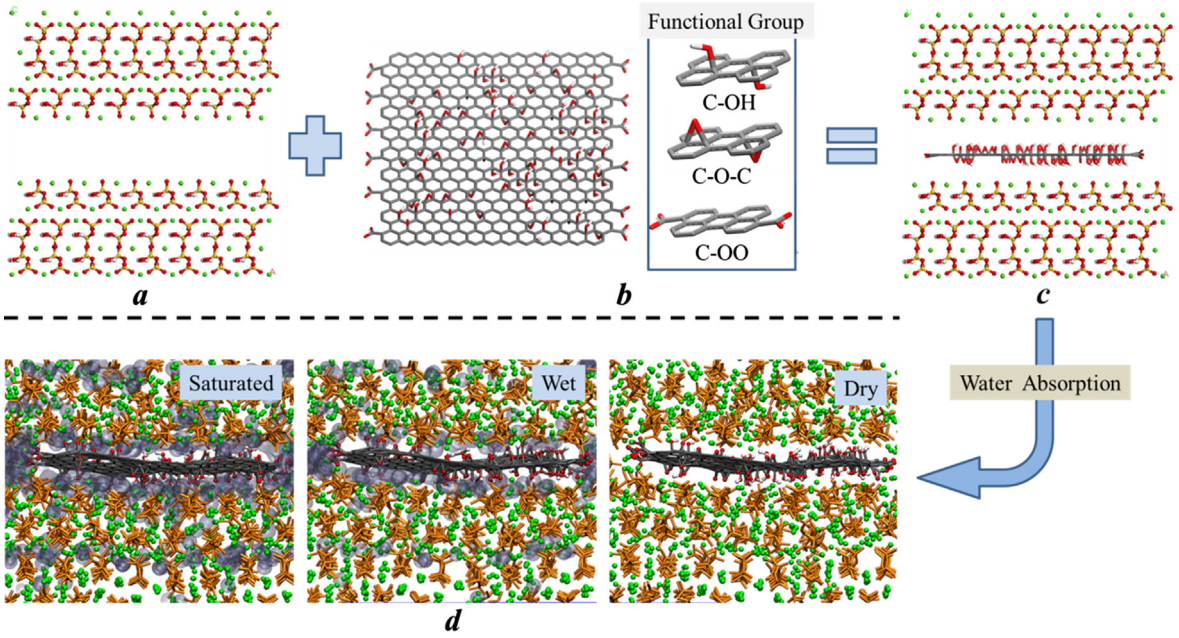


**Fig. 1.** Model construction of GO/C—S—H composite. Part *a* is cleaved tobermorite; part *b* contains GO sheet and functional groups; part *d* contains three GO/C—S—H composite models with different humidity states. The red, yellow, green, and white balls represent oxygen, silicon, calcium, and hydrogen, respectively. Yellow stick, gray stick, and gray bubble represent silicate bond, C—C bond, and water, respectively.

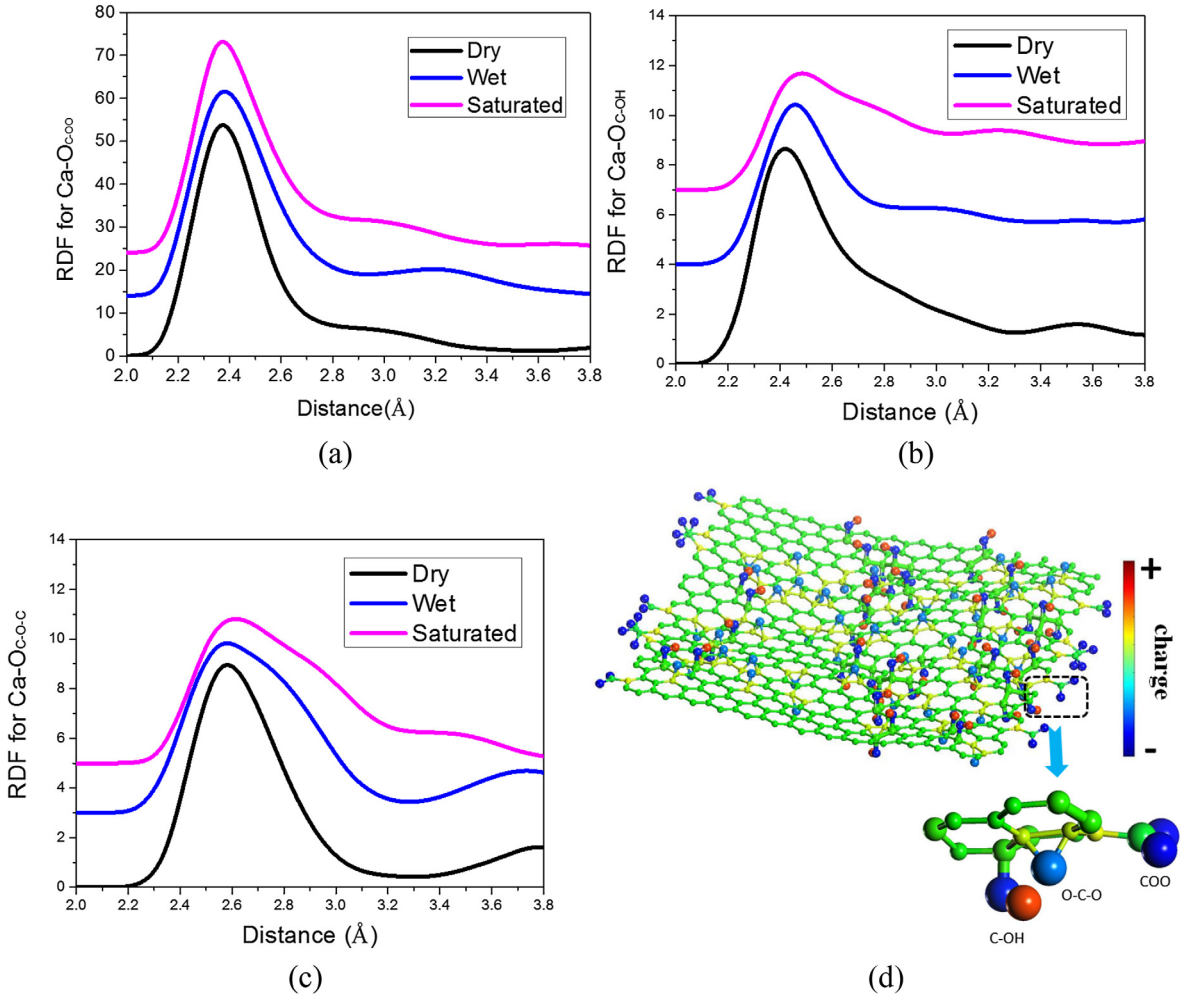


**Fig. 2.** Radial distribution function (RDF) of interlayer calcium associated with (a) $O_{COO}$, (b) $O_{C-OH}$, and (c) $O_{C-O-C}$, respectively; (d) charge distribution on the GO sheet.