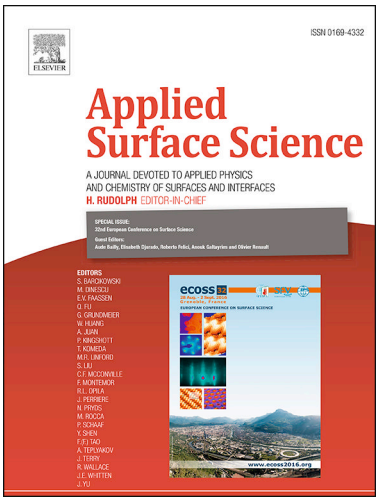

Full Length Article

Ionic hydration structure, dynamics and adsorption mechanism of sulfate and sodium ions in the surface of calcium silicate hydrate gel: a molecular dynamics study

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Ionic hydration structure, dynamics and adsorption mechanism of sulfate and sodium ions in the surface of calcium silicate hydrate gel: a molecular dynamics study

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Abstract: Molecular dynamics was utilized to investigate ionic structure, dynamics and adsorption behaviors of the sulfate and sodium ions highly solvated in the nanometer channel of calcium silicate hydrate (C-S-H) with two different Ca/Si ratios. Due to H-bond connection from silicate oxygen sites and attraction of surface calcium ions, a stable adsorption layer of water molecules was formed in the vicinity of the hydrophilic silicate channel: clear layered arrangements, strong orientation preferences and extremely low diffusion coefficients. Furthermore, the adsorption mechanisms differ for the sulfate and sodium ions confined in the gel pore of C-S-H: while sodium ions are connected onto the non-bridging oxygen sites by Na-O_{nb} bonding, the sulfate ions can be attracted by the calcium ions, leading to the aggregation of Ca-SO_4 ionic pairs. In the surface region, since the hydration shell of the solvated ions are partly substituted by the adsorbed water molecules, surface calcium ions and silicate oxygen sites, the diffusion of these ions is dramatically immobilized. Both the adsorbed Na^+ and SO_4^{2-} ions display extended exchange time for their solvation shells and longer residence time for their ionic clusters, as compared with those in aqueous solution. Besides, the increase in the surface Ca/Si ratio disturbs the local ordered arrangements and reduces the stability of the hydration layer. At high Ca/Si ratio, the highly solvated calcium ions contribute to the transformation from the outer adsorbed sulfate ions to the inner adsorbed species, enhancing the anions immobilization near the silicate surface.

Keywords: Molecular dynamics, C-S-H gel, Ca/Si ratio, sodium sulfate, structural and dynamical properties

1 Introduction

The transport and adsorption behavior of ions in the nanometer channel is of great importance in diverse scientific and industrial fields, such as batteries (Park, et al., 2010), building materials (Gollop & Taylor, 1992) and plant culture (Steudle, 2000). However, due to the impact of pore geometry (Kalinichev & Kirkpatrick, 2002), charge affinity and polarity of the substrate (Shirono & Daiguji, 2006), the confined ions differ dramatically from ones in the bulk solution in the respects of hydration structure, dynamic properties and hydrogen bonding (Wang, et al., 2004) (Hou, et al., 2015). In spite of great efforts on investigating the properties of confined