The properties of hydrophobic concrete prepared by biomimetic mineralization method

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Abstract. In this study, the calcium hydroxide, an inherent product of cement hydration, was treated using biomimetic carbonation method of incorporating stearic acid to generate the hydrophobic calcium carbonate on concrete surface. Carbonation reaction was carried out at various CO_2 pressure and temperatures and utilizing the SEM, chloride-ion penetration test apparatus, and compression test machine to investigate the hydrophobicity, durability, and mechanical properties of the synthesized products. Experimental results indicate that the calcium stearate may change the surface property of concrete from hydrophilicity to hydrophobicity. Increasing reaction temperature can change the particles from irregular shapes to needle-rod structures with increased shear stress and thus favorable to hydrophobicity and microhardness. The contact angle against water for the concrete surface was found to increase with increasing CO_2 pressure and temperature, and reached to an optimum value at around 90°C. The maximum water contact angle of 128.7 degree was obtained at the CO_2 pressure of 2 atm and temperature of 90°C. It was also found that biomimetic carbonation increased the permeability, acid resistance and chloride-ion permeability of the concrete material. These unique results demonstrate that the needle-rod structures of $CaCO_3$ synthetized on concrete surface could enhance hydrophobicity, durability, and mechanical properties of concrete.

Keywords: concrete; biomimetic carbonation; hydrophobicity; durability; strength

1. Introduction

Concrete is the most important and common construction materials used in civil engineering projects. It is essential that concrete is a durable material, that should withstand the conditions for which it has been designed, without deterioration, over a period of years (Deilami 2017). Devi (2018) indicated that durability is the property of concrete, which has the ability to withstand weathering action, chemical attack, abrasion or any other process of deterioration.

Service life of concrete depends mainly on its durability, i.e. the ability to resist physical

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effects (freezing and thawing, abrasion, erosion, drying shrinkage), chemical attacks (leaching and efflorescence, sulfate attack, acidic solution), weathering, and attack by natural or industrial liquids and gases. Li *et al.* (2017) and Li *et al.* (2018) have advised that the permeability of concrete is believed as the key property related to the serviceability and durability of concrete structures (e.g., bridges, hydraulic structures and marine structures) subjected to aggressive environments.

To improve the permeability of concrete, one of effective methods is waterproofing treatment which is adequate to make a hydrophobicity surface on concrete. Hydrophobic treatment can prevent wetting of the concrete's porous structure and in turn provides the self-cleaning ability that was called the "lotus effect" (Zhang *et al.* 2016). From the previous studies (Liu and Hansen 2016, Arabzadeh 2017, Sadowski and Stefaniuk 2018), making a hydrophobic surface has two requirements: rough surface and low surface energy. There are two methods could be produced a hydrophobic surface. First mothed, make a rough surface with a hydrophobic material, and second method is chemically modified the surface or coating a hydrophobic material upon it. To chemically modify the surface with a hydrophobic coating has several ways including sol-gel (Mishchenko 2012), dip coating (Ebert and Bhushan 2012), electrochemical (Zhang *et al.* 2004), and chemical or physical vapor deposition (Lau *et al.* 2003 and Hosono *et al.* 2005).

In addition, the hydrophobicity of concrete can be increased by forming a film of calcium carbonate to block the pores and reducing the permeability of the surface layer. Ca(OH)₂ can also be fixed by treatment with diluted water-glass. Calcium carbonate is then formed and filled the pores (Amidi and Wang 2015, Lertwattanaruk 2018). Treatment with magnesium fluorosilicate is also possible (Jia 2015 and Pana 2018). Concrete carbonation is a process in which carbon dioxide reacts with calcium hydroxide $(Ca(OH)_2)$ in the cement matrix to form calcium carbonate (CaCO₃) (Park 2008). Preparation of $CaCO_3$ in the presence of all kinds of organic substrate using soluble carbonate and calcium salts as initial materials has been reported by Wada 2004, Tong 2004 and Grassmann, and Löbmann 2004. It indicated that the organic substrate induced the nucleation and growth of CaCO₃, but does not change the surface property of CaCO₃. Keum et al. (2002) prepared hydrophobic CaCO₃ particles by mineralization with sodium trisilanolate in methanol solution. Chen et al. (2010) have reported carbonation of Ca(OH)₂ to synthesize cubic CaCO₃ using dodecanoic acid as modifier. Several organic additives used for synthesizing hydrophobic CaCO₃ via carbonation of Ca(OH)₂ include oleic acid (Wang and Sheng 2006), octadecyl dihydrogen phosphate (Wang and Xiao 2006), sodium oleate (Sheng 2006), stearic acid (Wang and Sheng 2007), and dodecanoic acid (Wang 2010).

In this research, the hydrophobic $CaCO_3$ nanoparticles were synthesized in situ in the presence of stearic acid by carbonating route via, mimicking the essential functions of bio-mineralization. The level of temperature and pressurized CO_2 of carbonation reaction was found to have a great effect on the morphology of $CaCO_3$ and thus the hydrophobic of the concrete surface. Furthermore, the carbonation and mineralization could not only induce the nucleation and growth of $CaCO_3$ nanoparticle but also improve the surface hydrophobicity, durability, and the compressive strength of the concrete.