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Technical note

Influence of cocamidopropyl betaine on the formation and carbonation of portlandite – A microscopy study

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ABSTRACT

In this study, the influence of concentrated cocamidopropyl betaine (TEGO® Betain F50) on the formation of calcium hydroxide and its carbonation is shown. Investigations to improve the comprehension of the impact on the morphologies and course of carbonation in the presence of the surfactant have been carried out, using several microscopic methods. In situ light microscopy was used to follow the carbonation reaction step by step. To get a closer view to the appearance of reaction products, fluorescence microscopy and electron microscopy has been applied. With the help of micro Raman spectroscopy adsorbed surfactant on the surface of portlandite crystals could be located. This work has been done to get a better understanding of the observations made in previous studies in the context of foam concrete and its optimization, using cocamidopropyl betaine as foaming agent.

1. Introduction

For the application of construction materials, different demands are made and properties are necessary. To adapt rheology, setting time and workability on the requirements and circumstances, organic additives like retarders, accelerators and superplasticizer are applied, for example [1]. In case of foam concrete, foaming agents are used to reach a certain air pore content and enable thermal insulation properties [2]. All additives affect the hydration process and their impact on cement hydration is strongly dependent on the present functional groups. Hereby, the additives can sorb on the surface of cement grains, form hydration products/seeds, complex calcium in the pore solution or build hardly soluble precipitates [3,4]. In consequence, the hydration course changes and the observed products may differ in their appearance and properties. In cement hydration calcium-silicate-hydrates (C-S-H) and calcium hydroxide (CH) are the main hydration products and both are influenced by organic additives but also other reaction products like ettringite are affected [1,3]. By influencing the growth mechanism and rate, crystal morphology is modified and this is strongly related to the concentration as well as the nature of the used additive. Present crystal planes are associated to the velocity in parallel shift. Planes with the smallest velocity remain whereby those with high velocity disappear. Surfactants can change these conditions, the resulting habitus and thus the crystal morphology. A change in physical–chemical parameters during crystallization, already during seed formation, takes place and growth centers are influenced [3]. Additionally, surfactants can be implemented as micelles in crystals and lead to structural and mechanical properties similar to biominerals. In sum, soluble molecules can influence and take part in both nucleation and growth [5].

Based on different studies it was found that cocamidopropyl betaine (betaine) has an influence on the pore structure of foam concrete and the formation of hydration products like CH and C-S-H phases. Also, the carbonation of CH is affected [6–9]. First insights into the influence of betaine on the formation of portlandite during diffusion synthesis and also the carbonation process were published [7,8]. Hereby, the authors showed that betaine chelates/adsorbs calcium ions, precipitates CH and possibly forms hydrotalcite-like reaction products, probably promoted in the presence of CO_2. Calcium hydroxide and –carbonate (Cc) were X-ray amorphous and both show smaller particle sizes and changes in morphology in the presence of betaine.

In this paper the influence of the surfactant, used in prior studies, on the formation of CH and its carbonation is focused more in detail to get closer insights and a better understanding on the observed pore structure as well as previous findings.

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2. Experimental section

2.1. Materials

As surfactant concentrated cocamidopropyl betaine (TEGO® Betain F 50, Evonik) and for synthesis of CH, KOH and $CaCl_2$ (Sigma Aldrich and Merck) were used.

2.2. Synthesis methods

Formation of CH by the means of diffusion crystallization was done by using KOH and $CaCl_2$ in a molar ratio of 2:1. Precipitation synthesis was carried out by dropping 20 mL 0.1 M $CaCl_2$ solution in 100 mL 1 M KOH solution using argon flow. For both synthesis solutions without (reference) and with 0.04, 0.4, 1.6 and 3.1% w/w betaine were prepared and investigated. Concentrations refer to the solution of surfactant in deionized water.

2.3. In situ microscopy

The used microscope is an Olympus BX 61, equipped for automatic treatment of samples with a 3D moveable stage with high precision, a connected digital camera, up to 500x optical magnification and sufficient resolution. Software for controlling the microscope is Analysis Pro. To record videos of reactions processes either every minute, in some cases every second minute, or every 30 s images have been taken. Due to the evaporation of water there is a focal shift during the measurements. Automatic focusing was programmed and to minimize the external impact, the aperture was programmed too, switching on the light only for taking images. The experiments took place under controlled and logged climatic conditions.

3. Results and discussion

3.1. Influence on the crystallization of $Ca(OH)_2$

To investigate the influence of different betaine concentrations on the morphology of CH, fluorescence microscopy (FM) and scanning electron microscopy (SEM) of formed crystals after diffusion and precipitation synthesis was done (Fig. 1).

The reference showed the typical morphology (habitus and tracht) of hexagonal oblong crystals. Compared to this, the crystals in presence of 0.04 and 0.4% w/w betaine were smaller with a rough

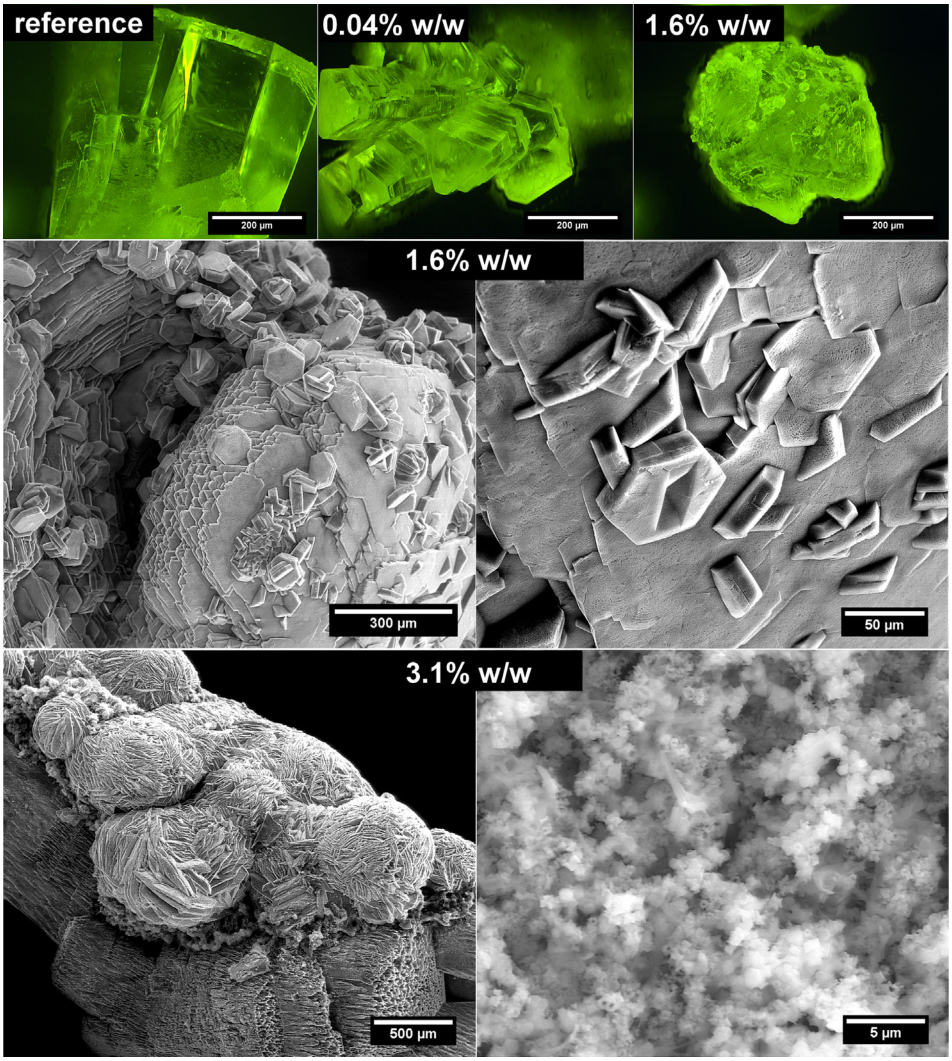

Fig. 1. Observed portlandite crystals by diffusion synthesis (upper left), adding 0.04% w/w (upper middle) and 1.6% w/w betaine (upper right) after one month by FM; Crystal growth in presence of 1.6% w/w (middle left and right), further growth of portlandite on a CH seed crystal in presence of 3.1% w/w betaine (lower left) and precipitation of CH with 3.1% w/w betaine (lower right) by SEM.