

Insights into the mechanisms of portlandite (Ca(OH)₂) sulfation

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In order to assess the mechanisms of hydrated lime (Ca(OH)₂, portlandite) reaction with SO₂ and to better understand the yield and kinetics of this reaction, which is of relevance for flue gas treatment applications, single crystals of portlandite were reacted with SO₂ vapours in a humid atmosphere at room temperature for different periods of time (1 to 28 days). The physicochemical, compositional and microstructural features of the SO₂-reacted portlandite single crystals were characterized using field emission scanning electron microscopy coupled with microanalysis via X-Ray fluorescence spectrometry (FESEM-EDS), transmission electron microscopy (TEM) along with selected area electron diffraction (SAED) and analytical electron microscopy (AEM), X-ray microdiffraction (m-XRD) and X-ray computed microtomography (m-CT). Sulfation of portlandite in contact with SO₂-rich atmosphere at a RH of 60 % results in the formation of pseudomorphs that preserve the external shape and volume of the original portlandite single crystals. Our observations suggest that portlandite sulfation occurs by a coupled dissolution–precipitation mechanism. The interaction between SO₂ and Ca(OH)₂ in the presence of adsorbed water results in the dissolution of the substrate (as seen by the formation of pseudo-hexagonal etch pits, with edges parallel to {100} directions) coupled with the precipitation of different Ca-bearing sulfite and sulfate phases, including amorphous precursors, on portlandite surfaces. Interestingly, despite the widely reported limited SO₂ removal performance achieved by this strategy due to the formation of a layer of newly formed phases covering the surface of portlandite crystals and hampering the diffusion of SO₂ towards the core of the crystal, we show that full replacement of the single crystals is achieved in our experiments. These (apparent) contradictory results are discussed in terms water availability in the system.