Calcite dissolution in claystones treated with brine and hydrogen: implications for underground hydrogen storage caprock integrity

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Underground hydrogen storage (UHS) in depleted oil and gas reservoirs or deep saline aquifers raises questions regarding changes to potential caprocks/seals that come into long-term contact with hydrogen. In this study we present the first results from a series of hydrogen treatments applied to caprock-analog claystones collected from quarries in Germany. One of four treatment options was applied to each individual sample: (i) untreated (reference), (ii) hydrogen gas treated (dry-H_2), (iii) NaCl brine treated (brine), and (iv) NaCl brine and hydrogen treated (brine-H_2). Although the dry-H_2 treatment option is unlikely to be reflective of in-situ UHS conditions, due to the likelihood of some fine-grained sedimentary rocks to swell and slake when saturated, the dry treatment option is included here to determine if any meaningful results may be obtained by the experimentally easier dry-H_2 treatment option. Following 30-day static treatments the samples were analysed via broad ion beam-scanning electron microscopy (BIB-SEM). From the SEM images, significant dissolution of calcite fossil fragments was observed in samples that were treated with a combination of hydrogen and 10 wt.-% NaCl brine. No significant textural changes were observed in samples that were treated with hydrogen alone. The initial results indicate that there is potential for alteration of calcite within caprocks of hydrogen storage systems, which could have consequences for long-term storage. Furthermore, the lack of dissolution observed with the dry hydrogen treatment indicates that, although experimentally easier, this treatment option does not result in significant dissolution. These results also highlight the need for further, robust testing of seals and caprocks in potential UHS systems.