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Impact of Mechanical and Chemical Alteration on CO_2 Leakage at the Cement-Casing Interface

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Possible CO_2 leakage through fractures in abandoned or improperly cemented wellbores is of concern for carbon capture and storage (CCS) projects. There is a large body of literature concerning steel degradation in presence of various fluids and gases. Many recent studies have also focused on the stability of cement in presence of CO_2. However, leakage risk in wellbores depends on the stability of both of these materials within the larger brine-CO_2-cement-steel system. The number of experimental studies focusing on flow within these multi-phase systems is considerably smaller. This work adds to the body of CCS-relevant knowledge by quantifying chemical changes in cement and steel materials upon exposure to CO2-equilibrated brine. Four cement-steel core samples were created with an artificial fracture between the two materials and were exposed to CO_2-equilibrated brine at various rates from 0.05 to 0.5 mL min^{-1}. To isolate the nature and extent of chemical reactions specific to each material, two additional experiments were conducted, using cement-cement and steel-steel samples with similar fracture surfaces, at a shared rate as one of the cement-steel experiments (0.1 mL min^{-1}). Changes to cement and steel fracture surface geometry, solid- and fluid-phase chemistry, and material properties were quantified via a combination of non-destructive characterization techniques (surface profilometry, X-ray tomography, solution chemistry analyses, and digital image correlation). Extensive reaction, inferred from elevated releases of iron and sulfur to solution and post-reaction changes to the steel surfaces, resulted in a constricted fluid pathway for the steel-steel sample. Conversely, changes to solution chemistry in cement-steel experiments were muted due to cement presence. When cement and steel surfaces were exposed to CO_2-equilibrated brine, pH levels were lowered, and metal release decreased compared to the steel-steel sample. Among the cement-steel experiments, slower flow rates resulted in the development of preferential reaction pathways and permeability increases, evident in both cement and steel portions of each sample. Faster flow rates tended to produce more homogeneous surface reactions along the fractures. Based on these results, we discuss implications for long-term behavior of casing-cement interfaces and the relationship between residence time and fracture sealing. Prepared by LLNL under Contract DE-AC52-07NA27344. LLNL-ABS-819782.