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Observational evidence for the long-term integrity of CO_2-reservoir caprocks

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Storage of anthropogenic CO_2 in geological formations relies on impermeable caprocks as the primary seal preventing buoyant super-critical CO_2 escaping upwards. Although natural CO_2 reservoirs demonstrate that CO_2 may be stored safely for millions of years, uncertainty remains in predicting how caprocks will react in contact with acid CO_2-bearing brines. This uncertainty is a significant barrier to risk assessment and consequently implementation of carbon capture and storage schemes. Prediction of caprock behavior is based primarily on theoretical modelling and laboratory experiments. However, the coupled reactive transport phenomena cannot be faithfully reproduced in laboratory experiments over sufficient timescales, theoretical models have not been calibrated against observational data and existing studies on natural caprocks have not resolved mineral reactions. Here we report the first detailed description and interpretation of a CO_2 reservoir-caprock system exposed to CO_2 over $\sim 10^5$ years, a time-scale comparable with that needed for effective geological carbon storage. Fluid-mineral reactions in the basal seven cm of the caprock, driven by diffusion of CO_2 and minor H_2S from the underlying reservoir, result in dissolution of haematite, dolomite and K-feldspar and precipitation of Fe-bearing dolomites, gypsum, pyrite and illite. The mineral dissolution reactions within the caprock generate transient increases in porosity but the propagation of these mineral reaction fronts is retarded by the reaction stoichiometry and mineral precipitation. Neutron scattering measurements indicate that the decrease in tortuosity and the fractal dimensions of the pore-network following mineral dissolution is only partly offset by mineral precipitation, implying a non-recoverable increase in effective diffusivity. Analytical modeling is used to extract kinetic data from the geometry of the mineral reaction fronts and numerical reactive transport modeling is used to place constraints on the time-scales of the alteration. The results attest to the significance of transport-limited reactions to the long-term integrity of sealing behavior in caprocks exposed to CO_2.