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Insights into vitrified nuclear waste alteration from glasses exposed to complex natural environments.

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Glass dissolution mechanisms have been well described as far as they relate to nuclear waste type silicate or borosilicate glass exposed to simple solutions, however, with countries now engaged in site selection for near surface or deep geological disposal facilities, there is increased urgency to understand glass alteration mechanisms in more complex systems. Recent studies of glass alteration in natural environments include those retrieved from shipwrecks, hyperalkaline caves, terrestrial sediments and replica burial mounds. Studying glasses from this range of natural systems, and comparing their alteration to that observed in short-term laboratory tests, highlights some common observations.

Studies demonstrate the role that external elements, sourced from groundwater and surrounding mineralogy, play in secondary phase formation. In particular, it was found that the sequestration of Ca and P over time resulted calcium-phosphate phases rich in heavy metals sourced from within the glass (for example Pb and the rare earth elements). Similarly, Fe was found to accumulate over time and associate with Si and Al. The two chemically distinct regions alternated to form bands and there was evidence for ongoing evolution of the alteration layer structure due to continued fluid ingress. Microorganisms were shown to affect glass alteration indirectly by controlling the local chemical environment, particularly the pH, the speciation of Fe and the availability of P. Results from unique field experiments are presented and compared with identical glasses altered under controlled laboratory conditions. Laboratory comparisons highlight the impact of surface finish on initial alteration rate and the importance of considering minor elements present at low concentrations that have the potential to sequester over time and have a major effect in the long-term.

Understanding glass dissolution and secondary mineral phase formation is key to predicting the long term retention of radionuclides and building the safety case for geological disposal of vitrified radioactive waste.