NMR studies of the impact of mineral transformation and sorbate aging on contaminant speciation and mobility

C. STREPKA[1], S. CHOI[2], P. O'DAY[2], J. CHOROVER[3] AND K. MUELLER[1]*

[1]The Pennsylvania State Univ., State College, PA 16802 (*correspondence: ktm2@psu.edu) (crs274@psu.edu)
[2]Univ. of California, Merced, CA, 95343 (poday@ucmerced.edu)
[3]University of Arizona, Tuscon, AZ, 85721 (Chorover@cals.arizona.edu)

Using a combination of analytical spectroscopy and wet chemistry techniques, we are developing a predictive-mechanistic understanding of the coupling between mineral weathering from caustic waste release and contaminant fate and transport in waste-impacted sediments from the Hanford DOE site. In these studies we have examined sets of samples ranging from speciman clays and homogeneous precipitates to Hanford sediments, each reacted with or formed from a simulated tank waste leachate (STWL) containing varying contaminant concentrations of Sr and/or Cs. Batch laboratory experiments as well as column studies have been performed, where total reaction times vary from 1 d to 1 yr.

Here we highlight synergistic results arising from the use of our main analytical research tool, solid-state nuclear magnetic resonance (NMR), along with supporting information from extended X-ray fine structure (EXAFS) spectroscopy. In this work, ^{29}Si and variable-field ^{27}Al magic-angle spinning (MAS) and multiple-quantum magic-angle spinning (MQMAS) NMR experiments provide kinetic data that follow the removal of octahedral aluminum species and the formation of neophases containing tetrahedrally-coordinated aluminum. Studies comparing homogenous precipitates from controlled model systems and more complex Hanford sediment samples reacted under similar conditions lend new insight into the molecular structures and kinetics of neoformed phases that sequester contaminants. Neophase formation is rapid in homogeneous systems (hours to days), with product phases influenced by the Si to Al ratio and the concentration and type of contaminant cation (Cs^+ or Sr^{2+}). For Hanford sediments, the rate of neophase formation is slower (days to months). Aluminosilicate neophases formed within one month are poorly crystalline and easily extracted, with EXAFS indicating Sr incorporation into cation sites in feldspathoid-type phases. Preliminary Sr and Cs NMR studies have also been conducted, and progress will be reported here.

The mode of plume-ridge interaction: Constraints from He, Ne, Ar isotope and abundance systematics

N.A. STRONCIK[1], S. NIEDERMANN[1], K. HAASE[2], K. HOERNLE[3], F. HAUFF[3] AND R. WERNER[1,2,3]

[1]GeoForschungsZentrum Potsdam, Telegrafenberg, D-14473 Potsdam, Germany
[2]GeoZentrum Nordbayern, Schlossgarten 5, D-91054 Erlangen, Germany
[3]IFM-GEOMAR, Wischhofstr. 1-3, D-24148 Kiel, Germany

In areas where hotspots are located in the vicinity of mid-ocean ridges (MORs), the deep mantle plume flow creating the hotspot at the surface interacts with the ridge system, as indicated by along-axis bathymetric and gravitational anomalies as well as by geochemical and isotopic anomalies of mid-ocean ridge basalts (MORBs). Today, 21 of the about 50 active hotspots have been identified to interact with MORs, inducing such anomalies along 15 to 20% of the length of the global MOR system. Despite the importance for plate production the actual mode of plume-ridge interaction remains enigmatic. One of the main outstanding problems in this context is the physical state in which mantle flow and mixing occur in those settings. Mixing of plume mantle and MORB source material may either be in the form of two solids, of solid-melt or of two melt phases only. Based on the first noble gas isotope and abundance data from the Foundation Seamount Chain/Pacific-Antarctic-Rise system and from the Galápagos spreading centre and adjacent off-axis seamounts we show that mixing between the plume and the MOR material occurs in the physical form of melts. The positive correlation of ^{4}He/^{40}Ar* with ^{206}Pb/^{204}Pb as well as the He and Ne versus Pb isotope systematics indicate that both the plume and the MOR source material have been affected by degassing and melting processes prior to mixing. However, the occurrence of e.g. degassing processes requires the existence of melts, implying that mixing between both mantle materials occurs in the physical form of melts.