

Linear trend of mid-continent alkaline volcanism, USA: slab-edge focus

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Alkaline volcanism in the mid-continent USA includes a linear zone of Cretaceous-Tertiary kimberlite-carbonatite magmatism. Magmatism along the N40°W linear trend from Louisiana to Alberta occurs at 109-85, 67-64, 55-52, and less than 50 Ma. Magmatism along the southern part of the trend occurred at 109-85 and 67-64 Ma, and magmatism in the north occurred during the last three pulses. In Arkansas, initial Sr-Nd-Hf-Pb isotopic compositions indicate that there were at least two distinct melt sources for alkaline rocks. One end member is characterized by low ⁸⁷Sr/⁸⁶Sr and high ¹⁴³Nd/¹⁴⁴Nd and ¹⁷⁶Hf/¹⁷⁷Hf, similar to values of oceanic intraplate basalts interpreted as originating by melting of convecting mantle sources. The other end of the isotopic arrays extends to higher ⁸⁷Sr/⁸⁶Sr, but most clearly lower ¹⁴³Nd/¹⁴⁴Nd and ¹⁷⁶Hf/¹⁷⁷Hf than seen in most intraplate oceanic basalts. In the north, there are three Sr-Nd isotopic groupings, with latest kimberlite-carbonatite volcanism having the lowest initial ⁸⁷Sr/⁸⁶Sr and highest initial ¹⁴³Nd/¹⁴⁴Nd ratios.

The position of the magmatic track roughly parallel to the western subduction margin of the North American plate, the lack of spatial age progressions of the magmatism that would be consistent with motion of North America over a fixed hot spot, and the presence of Sr-Nd-Hf-Pb isotopic and trace-element compositions that show a temporal evolution from lithospheric to asthenospheric melt-sources, suggest that this linear zone is the surface expression of mantle melting somehow connected to the behavior of the subduction system ~2000 km to the west. We propose that fragmentation of the Farallon and Kula plates, as imaged by tomography beneath North America, opened slab windows parallel to the trench, but well to the east of the convergent margin. These induced sheet-like mantle upwelling along their margins, resulting in small-degree decompression melting that produced highly alkaline magmas.

Tin sorption to magnetite nanoparticles under anoxic conditions

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The long-lived fission product ¹²⁶Sn is of substantial interest in the context of nuclear waste disposal in deep underground repositories. However, the redox state (di- or tetravalent) under the expected anoxic conditions is still a matter of debate. We therefore investigated sorption and oxidation of Sn(II) in the presence of a typical corrosion product, magnetite (Fe^{II}Fe^{III}₂O₄), with a mean particle size of 9.4 nm. In order to simulate waste disposal conditions, the experiments were performed under strictly anoxic conditions in a glovebox at <2 ppm O₂. Macroscopic parameters (pH, Eh, [Sn], [Fe]) were monitored along with redox state and local structure of Sn (X-ray absorption spectroscopy) and Fe (XPS) as a function of time, pH, and surface loading.

Magnetite rapidly sorbed Sn(II), reducing Sn concentration within 0.5 h from 10 to 0.0084 μM. Tin was strongly sorbed by magnetite across a wide pH range from 3 to 9. Reduced sorption at pH <3 is in line with electrostatic repulsion between the positively charged surface of the magnetite nanoparticles (IEP ~6.7) and cationic Sn²⁺ or Sn⁴⁺ complexes. The reduced sorption at pH > 9 is in line with the transition from Sn(OH)₂⁰ to the anionic Sn(OH)₃⁻ which occurs at pH 9. Across the pH range 3-9 and reaction periods ≥1 h, EXAFS-derived sixfold oxygen coordination and XANES edge energy positions of ~29207 eV both indicate the presence of Sn(IV) at the magnetite surface. EXAFS shell fitting as well as Monte Carlo simulations showed formation of edge-sharing complexes of Sn(IV) with FeO₆ octahedra (Sn-Fe distance of 3.15 Å), and formation of corner-sharing complexes with FeO₄ tetrahedra (Sn-Fe distances of 3.60 Å). Even after the longest reaction periods of 1 month, we did not observe incorporation of Sn(IV) into the (compatible) magnetite structure. Also, precipitation of SnO₂ was not observed in spite of supersaturation.

In order to elucidate the reaction pathway, we also studied Fe in solution and at the surface (XPS). Starting with the PZC and increasing with [H⁺], the magnetite surface released Fe(II) into solution (0.11 g/L at pH 2). After addition of Sn(II), however, [Fe] in solution decreased as a function of Sn loading, in spite of the expected increase of structural Fe(II) due to the coupling to Sn(II) oxidation. This suggests a re-adsorption and possible re-precipitation of Fe(II) at the magnetite surface. Nevertheless, Fe(II) again re-dissolved as a function of time at low pH. With XPS we were not able to detect an adequate increase of the Fe(II)/Fe(III) ratio at the surface, supporting an electron redistribution between bulk and surface Fe centers.

In conclusion, our study demonstrates that Sn is strongly retained by magnetite across a wide pH range, forming stable surface complexes and stabilising the magnetite surface against dissolution.