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Oxidation of hydrogen sulfide by Fe(III) in phyllosilicates

Conference Paper · March 2002

DOI: 10.13140/RG.2.1.3754.6642 · Source: OAI

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The dynamics of melting beneath Theistareykir, northern Iceland

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U-Th disequilibrium data on a suite of 34 postglacial basalts (< 10,000years old) from Theistareykir, northern Iceland, show a relatively high degree of disequilibrium compared to "normal" mid ocean ridge basalts (N-MORB). For the bulk of the samples, ²³⁰Th excesses are within 25±5%, but both the most depleted samples ("picrites") and the geochemically and isotopically most enriched basalts from locations adjacent to Theistareykir (Draugarhraun and Asbyrgi) have lower ²³⁰Th excesses of about 15%. The large range in chemical and isotopic variations of the Theistareykir samples provides an excellent opportunity to investigate the influence of source heterogeneity on the degree of disequilibrium. The lack of correlation between U-Th data and radiogenic isotope or chemical parameters indicates that source heterogeneity does not play an important role in creating the $(^{230}\text{Th}/^{238}\text{U})$. Therefore, the Theistareykir basalts provide clear evidence that the Th systematics result from variations in the timing of the melting and melt extraction process only. The relatively large and constant ²³⁰Th excesses for the bulk of the samples (20-30%) suggest that melting and melt extraction must be relatively reproducible, with the melting starting well within the garnet-stability field and requiring residual porosity $\leq 0.1\%$, upwelling rates ≤ 1 cm/yr (corresponding to melting rates $\leq 1*10^{-4}$ kg/m³yr), and melt ascent velocities \geq 3-4m/yr. Lower ²³⁰Th excesses of about 15% in both the most depleted and enriched lavas indicate some variations in the melting and melt extraction process. The low ²³⁰Th excesses of the most depleted melts could either be an effect of slow melt migration, or be caused by incomplete melt aggregation, with melts created at the very bottom of the melting regime being underrepresented. The low $^{\rm 230} {\rm Th}$ excesses of the most enriched lavas could be due to slow melt migration for melts created at some distance from the center of the melting column and could also be influenced by radioactive decay during crystal fractionation in crustal magma chambers.

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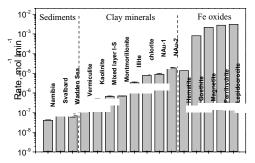
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Phyllosilicates (e.g., smectite and illite) in marine sediments may contain appreciable amounts of redox-active structural Fe(III). The role of clay-bound Fe in the oxidation of microbially produced hydrogen sulfide in marine sediments has not been yet evaluated.

Different Fe(III)-containing clays (montmorillonite SWy-2, nontronites NAu-1 and NAu-2, illite IMt-1, mixed layer illite-smectite ISCz-1, kaolinite KGa-2, chlorite CCa-2, and vermiculite) as well as the <2µm fraction of marine sediments (German Wadden Sea, fjords of Svalbard, and Atlantic Ocean off Namibia) were used for experiments after the removal of organic matter and Fe oxides.

Initial rates of H_2S oxidation by different minerals were obtained for environmentally important conditions (35% seawater, t = 20°C, pH = 6.0-8.5, H_2S_{init} = 60-200 μ M). The reaction rate was found to be highly pH-dependent for Fe-rich smectites (NAu-1 and NAu-2), but not for other clay minerals.

Figure 1: Rates of H₂S oxidation by different Fe(III)containing minerals.



The rates of H_2S oxidation (pH 7.0) for different clays vary within two orders of magnitude. Nontronite is capable of oxidizing H_2S at rate comparable to that of the least reactive iron oxide, hematite (dos Santos Afonso and Stumm, 1992) (Fig. 1). The rates of hydrogen sulfide oxidation is generally controlled by Fe(III) concentration in the clay mineral.

References

dos Santos Afonso, M. and Stumm, W. (1992) Langmuir, 8, 1671-1675.