

Lu-Hf and Sm-Nd systematics of the first solids in the Solar System

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Hafnium-176 excesses are found in bulk chondrite and eucrite isochrons, and in some (not all) eucrite [1] and angrite mineral isochrons [2,3]. It was proposed that cosmic rays accelerated by supernova(e) shocks in the early Solar System could produce a higher decay rate of a ¹⁷⁶Lu isomer within the irradiated planetary materials [2]. This hypothesis complicates the direct determination of the initial Lu-Hf isotopic composition of the Solar System and by extent that of the terrestrial planets [4]. The interpretation of the earliest terrestrial Lu-Hf isotopic records relies on knowing these initial conditions [5].

Such external irradiation sources should affect any objects already formed and exposed in the protoplanetary disk. We thus investigated the stable and radiogenic Lu-Hf and Sm-Nd compositions of CAIs, the first solids formed in the Solar System. The selected five CAIs were formed after injection of ²⁶Al into the protoplanetary disk [6]. The ¹⁴⁴Sm/¹⁵²Sm ratios have the largest variations with p-process deficits typically at ~250ppm. Deficits in ¹⁴⁹Sm (<100ppm) are correlated to excesses in ¹⁵⁰Sm within the same CAI suggesting small neutron capture effects. All stable Nd isotope ratios have deficits smaller than 50ppm. The bulk CAIs have no resolvable Hf isotopic variations for ¹⁷⁴Hf, small positive anomalies (<20 ppm) for ¹⁷⁸Hf and ¹⁸⁰Hf which demonstrate minimal corrections (<40ppm) for ¹⁷⁶Hf/¹⁷⁷Hf ratios. The bulk and mineral ¹⁴⁷Sm-¹⁴³Nd and ¹⁷⁶Lu-¹⁷⁶Hf isochron ages for three melted CAIs together are 4519 ±140 Ma (MSWD =0.77, ¹⁴³Nd/¹⁴⁴Nd_i = 0.50675 ±20) and 4560 ±190 Ma (MSWD =59, ¹⁷⁶Hf/¹⁷⁷Hf_i = 0.27985 ±15) respectively. We thus interpret the anomalous Lu-Hf isochron slopes [1,2] as the consequence of secondary parent body processes. Our direct determination of the initial ¹⁷⁶Hf/¹⁷⁷Hf of the Solar System is consistent with, but less precise than, those obtained from present CHUR values and from updated bulk chondrite isochrons [7].

[1]. Righter et al. (2013), 44th LPSC, A#2745. [2]. Thrane et al. (2010), ApJ **717**, 861-867. [3]. Sanborn et al. (2012), 43rd LPSC, A#2039. [4]. Bizzarro et al. (2012), *Geochem. Geophys. Geosyst.* **13**, Q03002. [5]. Harrison (2009), *Annu. Rev. Earth Planet. Sci.* **37**, 479-505. [6]. Krot et al. (2012), *MAPS* **47**, 1948-1979. [7]. Bouvier et al. (2008), *EPSL* **273**, 48-57.

Ion concentration at the kaolinite – water interface

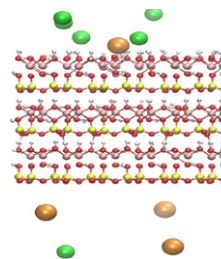
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Kaolinite is a clay present in soils, sediments and rocks, including sandstones and carbonates, as well as shales and mudstones. Its high surface area and large cation exchange capacity make it reactive in natural environments. For example, it plays an important role in uptake of contaminants in the environment and in tertiary oil recovery.

To observe the surface chemical composition of kaolinite, as it is during contact with solution, we used X-ray photoelectron spectroscopy (XPS) with the fast freezing technique. This allows the mineral - fluid interface to be preserved and investigated, even under ultrahigh vacuum which is necessary for XPS, because the ion distribution is “frozen in” as it was in the original liquid phase.

Solutions investigated were NaCl, CaCl₂ and Ca(C₂H₅COOH)₂ at pH 5.5, in their reaction with pure kaolinite (KGa1-b). The results show that the relative ion concentration is different at the surface than in the bulk, consistent with electrical double layer (EDL) theory. We also observed that the ratio of Ca and Cl ions is not constant with distance out from the interface. For low CaCl₂ concentrations (< 6 mM), the Ca:Cl ratio is higher at the interface, whereas at higher Ca concentration, more negative counter ions, i.e. Cl, are present to balance charge. Results from molecular dynamics (MD) simulations (figure) and atomic force microscopy (AFM) experiments complement the XPS results with structural information to build a full picture of interface interaction.



MD simulation snapshot of Ca (orange) and Cl (green) adsorbed on kaolinite during exposure to solution. Water is present but we have made it invisible.