New evidence for chemical fractionation of radioactive xenon precursors in fission chains: Geochemical implications

ALEX MESHIK* AND OLGA PRAVDIVTSEVA

Washington University, Saint Louis, USA
(am@wustl.edu)

Until recently the effect of chemical fractionation of radioactive Xe precursors in fission chains has been reliably observed only in samples irradiated in nuclear reactors, including natural reactor in Oklo operated in the pulsed mode. Here, for the first time, we experimentally demonstrate that apparent Xe fission yields can be significantly modified in common ~ 135 Ma U-bearing rocks which experienced relatively small and nearly constant natural neutron flux.

Sample was split into two aliquots, the first one analyzed as is, and the second one subjected to acid treatment which preferentially leached out U-bearing mineral phases but did not affect silicates. Xe from both aliquots has been extracted by step-wise pyrolysis and analyzed in the same way. Isotopic composition of Xe from the untreated sample was found to be consistent with ^{238}U fission spectrum with minor addition of Xe from neutron induced ^{235}U fission for all extraction steps. While isotopic composition of Xe from the chemically treated aliquot was indistinguishable from the composition of untreated sample at temperatures steps above 800°C, Xe released at lower temperatures was significantly depleted in heavy isotopes. The isotopic shifts, revealed here by acid treatment, correlate with half-lives of radioiodine, a volatile, water-soluble Xe precursor in fission chains (Fig. 1).

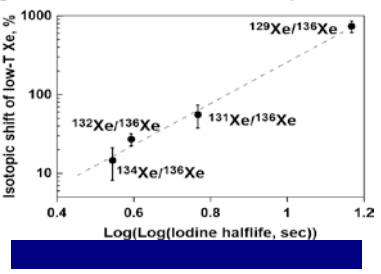

Fig.1. Isotopic shifts (normalized to ^{136}Xe) in fission Xe released below 800°C from the sample treated with HNO_3 *vs.* half-lives of radioiodine. Errors are 1σ.

This correlation (over 13 orders of magnitude in half-lives!) is caused by displacement of fission products from the radiation damage zones by distances depending on time available for the diffusion of their immediate precursors.

This isotopic effect will not be observable in bulk unaltered samples, but we expect to see it in rocks affected by aqueous alteration and/or weathering, processes that are common on Earth. Complimentary isotopic compositions then should contribute to Xe isotopic signature in natural gases and possibly in Earth's atmosphere.