ISOTOPIC FRACTIONATION OF FISSION XENON FROM TEMPERATURE VARIATIONS: IMPLICATIONS FOR THE OPERATIONAL CONDITIONS OF THE OKLO REACTOR.

A. P. Meshik, C. M. Hohenberg, O. V. Pravdivtseva. Laboratory for Space Sciences and Physics Department, Washington University, St. Louis, MO 63130, am@wuphys.wustl.edu

Introduction. Anomalous fission Xe, isotopically enriched in ^{132}Xe, ^{131}Xe and ^{129}Xe, was observed in Al-phosphate from Zone 13 of Oklo reactors [1]. However these anomalies have never been addressed quantitatively. Here we report new data and suggest a quantitative model for the observed isotopic effects (chemical fractionation). We use this model to estimate the operational conditions for this natural reactor.

Experimental. In addition to previously reported data [1, 2], we have analyzed a new polished section of material from Zone 13 with *in-situ* laser extraction. The Xe concentrations in U-free Al-phosphate ($\sim 10^{-2}$ cm^3 STP/g) were ~ 100 times higher than in U-oxides. Xe isotopic compositions in Al-phosphates were found to be extremely anomalous, while the Xe in U-oxides was relatively "normal" (mixtures of ^{235}U, ^{239}Pu and ^{238}U fission). To produce anomalies observed in Al-phosphates, Xe isotopes must have been generated in following proportions: $P_{132}/P_{134} = 8.0$; $P_{131}/P_{134} = 3.4$; and $P_{129}/P_{134} = 1.05$.

Model. The proposed model is based on three assumptions. (1) Natural reactors operate in pulse modes. During a single pulse the temperature increases until the moderating water vaporizes, terminating the chain reaction. Then the temperature slowly decreases, water returns back to the active zone and the chain reaction resumes. This negative feedback prevents runaway chain reactions after the natural reactor goes critical. (2) At elevated temperatures Te and I (radioactive Xe precursors in fission chains) are less mobile than Xe. Experimentally determined Te, I and Xe retention [*e.g.* 3] supports this assumption. (3) During the reactor operation fission Te and I migrate from U-oxide into Al-phosphate where they subsequently decay to Xe, which is only retained when the reactor cools down between pulses.

Operational conditions. We have calculated the isotopic composition of Te as a function of two variables: the duration of a typical pulse of the reactor (**d**) and its inter-pulse cooling time (c) when Xe retention begins. We find a good (and only) match between the model and the three Xe ratios observed in the Al-phosphates with the parameters **d** = 30 min and **c** = 2¾ hr, with **c** being a lower limit for the average time between the termination of a one pulse and the initiation of the next pulse. Interestingly, these parameters are quite typical for geysers (**c** = 5 to 50 minutes, **d** = ⅓ to 3 hours, [4]).

Implication. The proposed fractionation mechanism, and this model for Xe isotopic fractionation, may be useful not only in this study of natural reactors, but also in any process in which nuclear production involves intermediate products and takes place in an environment with varying temperature. As is this case, pseudo-components may be also generated by similar chemical migration.

References. [1] Meshik A.P., Kehm K., and Hohenberg C.M. (2000) *GCA* 64, No. 9, 1651–1661. [2] Meshik A.P., Hohenberg C. M., Pravdivtseva O.V. (2003) *Geophys. Res. Abstr.*, 5, 14271. [3] Curtis D., Benjamin T. Gancarz A. (1989) *Applied Geocheistry* 4, 49–62. [4] http://www.geyserstudy.org