

MICRO-DISTRIBUTION OF FISSION XENON ISOTOPES: A POSSIBLE EXPLANATION OF XENON COMPOSITION IN PHASE Q. A. Meshik, O. Pravdivtseva and C. Hohenberg. McDonnell Center for Space Sciences and Physics Department CB1105, Washington University, St. Louis, MO 63130 (am@physics.wustl.edu).

Introduction: Fission of heavy nuclei is energetic process causing several μm recoil of fission fragments and evidently could result in significant losses from submicron grains. Whether or not these losses are identical for all isotopes of fission products depend on the micro-distribution of fission fragments. Kennett and Thode [1] studied the micro-distribution of Xe isotopes in neutron irradiated U_3O_8 powder imbedded in U-free matrix and found that at low extraction temperatures the apparent fission yields of ^{131}Xe and ^{132}Xe in this material was enriched ~ 10 -fold relative to ^{134}Xe and ^{136}Xe . A modified version of their experiment included the isotopic analyzes of the Xe accumulated in the neutron irradiated quartz ampoule containing the U-oxide powder [2]. Fission Xe in the ampoule was found to be enriched in ^{136}Xe and ^{134}Xe , complimentary to the low temperature Xe releases from the irradiated powder itself which was depleted in these isotopes. In both experiments samples received high neutron dose in relatively short time which is not typical for natural environments, with the exception of the Oklo reactors in Gabon [3] which still remains the only occurrence of natural nuclear chain reaction. This isotopic modification of apparent fission yields was attributed to CFF-effect (Chemically Fractionation Fission) [4] and was considered to be a rather exotic process observed mainly in the environments with pulsed neutron flux or short neutron bursts. There were several observations of ^{131}Xe and ^{132}Xe enrichments, similar to those found in the above experiments, but they were either not reproducible [5, 6] or tentatively attributed to neutron capture reactions on Te [7]. The experiment reported here demonstrates that CFF process can modify apparent fission yields not only in the reactors and at nuclear test sites, but also in conventional U-bearing ores where the relatively low neutron flux is nearly constant over time and the temperature changes are slow.

Experiment: The sample used in this work was a fragment of a common uranium rich ore: an assemblage of various uranium oxides, brannerite, coffinite closely overgrown with silicates, forming a U-Si metagel as the result of secondary alteration of coffinite [8]. Xe release profile obtained during stepwise pyrolysis of the original sample (Fig. 1a) was a superposition of Xe released from different minerals constituting the assemblage. Isotopic composition of Xe (after subtraction of small atmospheric contamination) was uniform in all temperature steps.

The aliquot of the same sample was powdered to $<74 \mu\text{m}$ and treated with 6M HNO_3 at room temperature until the reaction completed. About 80 wt.% of the

original sample has been dissolved by this treatment. The release profile of fission Xe from the treated sample (Fig. 1b) revealed a noticeable low temperature release peak apparently absent in the original sample.

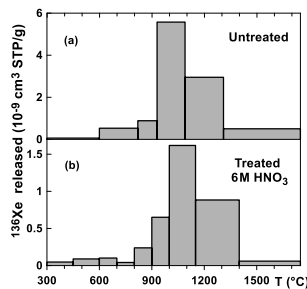


Fig. 1. Release of fission Xe from the original U-bearing sample (a) and from the sample treated with HNO_3 (b). Xe release below 800°C in the acid treated sample tend to form distinct peak with anomalous isotopic composition shown in Fig. 2.

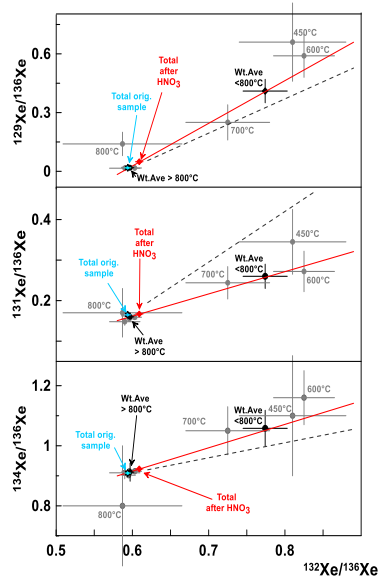


Fig. 2. Isotopic composition of Xe released from acid treated sample during stepped pyrolysis. All temperature fractions form linear arrays (solid lines) different from mass fractionation lines (dashed). Fission Xe in all temperature fraction of original sample (blue) is isotopically uniform. Xe in low temperature fractions of treated sample (black) are the different from fractions above 800°C and from the bulk composition (red).

The isotopic shifts of weighted average fission Xe released below 800°C from the bulk Xe composition are correlated with half-lives of radioiodine, an immediate Xe precursor (Fig. 3).

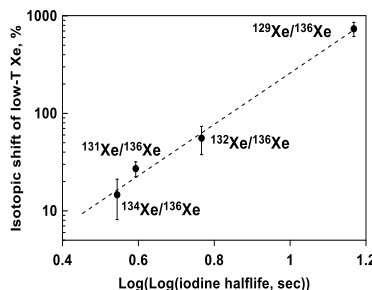


Fig. 3. Isotopic shifts in fission Xe (^{136}Xe normalized) released below 800°C from the acid treated U-bearing sample vs. half-lives of corresponding I isotopes, Xe precursors in fission chains.

This correlation (over 13 orders of magnitude in half-lives!) is evidently caused by higher mobility of chemically active radioiodine, a precursor of Xe in fission

chains. Apparently, the longer an I isotope lives before decay, the greater distance it can diffuse. After iodine decays, the resulting Xe isotope will reproduce the spatial distribution of its precursor since any β -recoil effect is negligible.

Our experiment has demonstrated that micro-distribution of fission Xe is isotope-specific even in conventional terrestrial rocks with uniform neutron flux. The acid attacks the radiation damaged zones first releasing the least retentive ^{136}Xe and ^{134}Xe , making the remaining fission Xe complementarily enriched in ^{132}Xe , ^{131}Xe and ^{129}Xe . This remaining Xe is released at low extraction temperatures, perhaps indicating association with near surface residences and/or the association with the smallest grains. This implies that CFF process works only on a small scale and must be averaged on a large scale, in the bulk rock. Indeed in the untreated sample we do not observe any statistically significant isotopic effects.

Both aqueous alteration and weathering reduce effective grain sizes increasing the surface areas and exposing radiation-damaged zones. In this case partial fission xenon losses could be accompanied with isotopic modification caused by the CFF-effects. This process is apparently at work at the DOE's Hanford Site in SW Washington State. Xe releasing from highly radioactive man-made transuranic waste is enriched in the least retentive ^{136}Xe and ^{134}Xe , with the former being approximately twice higher than the latter [9].

Xenon in phase Q: Phase-Q contains the majority of Xe in primitive meteorites. Isotopically Xe-Q is very close to solar Xe fractionated by $\sim 1\%/u$ with 1.5% addition of Xe-HL apparently produced by in supernovae and associated with diamond-rich separates from meteorites [10]. It was recently suggested that in addition to carbonaceous carrier phase Q may contain sulfides known to host U, and therefore Xe-Q can be modeled as fractionated solar Xe with the addition of uranogenic Xe. However, this model does not pass the likelihood criterion: the difference between Xe-Q and the model composition exceeds 7σ for ^{132}Xe and ^{134}Xe . Meanwhile the model composition involving Xe-HL agrees with the Xe-Q within 1-2 σ [10]. The association of solar and "presolar" components in phase Q does not have a satisfactory explanation and it is difficult to explain why a nanodiamond-free Saratov (L4) has high concentration of Xe-Q [12].

Phase-Q has a clear excess of ^{129}Xe from extant ^{129}I (15.7 Myr), but apparently little, if any, fission Xe from ^{244}Pu (80 Myr). Our experiment suggests that, due to CFF process, a micro-distribution of fission Xe is not uniform, but isotope-specific. Phase-Q is very fine-grained substance separated by severe chemical treatment, somewhat similar to what we applied to the U-bearing sample in our experiment. We therefore may

expect that CFF-process could be at work there and it could modify apparent fission yields from ^{244}Pu fission in the same manner as our experiment modified fission yields of ^{238}U .

Figure 5a shows the composition of Xe-Q fractionated by 0.65%/u in order to match fission-free $^{124-128}\text{Xe}$ isotopes to solar Xe. After this procedure $^{131-136}\text{Xe}$ become overabundant comparing to the solar Xe. The composition of these isotopes does not match ^{244}Pu or other known fission spectra. But it match very well the composition of Xe releasing from the Hanford nuclear waste site (Fig 5b). Therefore phase-Q may contain Pu-fission Xe, not a normal one, but modified by CFF in the same way as Xe in the Hanford site was modified from normal ^{235}U fission Xe. Having completely different nature and origin, these two objects may have one thing in common – they selectively captured least retentive CFF-Xe isotopes, from ^{235}U in Hanford site and from ^{244}Pu in phase Q. Therefore we can model composition of Xe-Q as 0.65%/u fractionated Solar Xe with 3% (for ^{136}Xe) addition of CFF-Xe from ^{244}Pu .

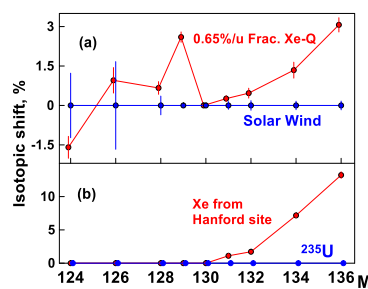


Fig. 5. Xe composition of fractionated Xe-Q matches Xe releasing from nuclear waste facility. The only exception is ^{129}Xe which did not have enough time to accumulate from decay of fissionogenic ^{129}I at the Hanford site.

Conclusion: CFF is not yet another Xe component with a unique isotopic composition, but is a physiochemical process modifying micro-distribution of fission Xe. It may explain the composition of Xe-Q without involving pre-solar components.

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[1] Kennett T. J. and Thode H. G. (1960, *Can. Journ. Phys.* 38, 945. [2] Shukolyukov Yu. A. et al (1994) *Geochim. Cosmochim. Acta* 58, 3075–3092. [3] Meshik A., et al (2004) *Phys. Rev. Lett.* 93, 182302-1 – 182302-4. [4] Meshik A. (1988) *PhD Thesis*, Vernadsky Institute, Moscow. [5] Jeffery P. M. (1971) *Nature* 233, 260 –261. [6] Azuma S., et al (1993) *Earth Planet. Sci. Lett* 114, 341–352. [7] Drescher J., et al (1998) *Earth Planet. Sci. Lett.*, 154, 247–263. [8] Aleshin A. (2008) *PhD Thesis*, de l'Université Henry Poincaré, Nancy-I. [9] Dresel P. E. et al (2008), *Journ. Radioanal. Nucl. Chem.* 276, 763–769. [10] Gilmour J. D. (2010) *Geochim. Cosmochim. Acta* 74, 380–393. [11] Marrocchi Y. et al (2015) *Geophys. Res. Lett.* 2093–2099. [12] Amari S. and Meshik A. (2015) *Met. Soc. Meeting*, Abstract #5213.