On the origin of Xe-isotope anomalies: A new approach based on the ^{131}Xe/^{134}Xe vs ^{132}Xe/^{129}Xe diagrams

S. D. Mineev — Laboratory of Isotope Geochemistry, Vernadsky Institute of RAS, Moscow, 117975, Russia

V. V. Poukhov — Department of Geology, Moscow State University, Moscow, 117234, V-234, Russia

V. B. Polyakov
A. M. Pleshakov — Laboratory of Isotope Geochemistry, Vernadsky Institute of RAS, Moscow, 117975, Russia

E. K. Jessberger — Institut für Planetologie, Westfälische-Wilhelms-Universität, Wihelm-Klemm-Str. 10, 48149, Münster, Germany

The previous discussion by Shukolyukov *et al.*, (1994) of the origin of anomalous Xe isotope compositions that were observed in natural gases, meteorites, and the Oklo uranium deposit did not include the ^{128}Xe, ^{130}Xe and ^{136}Xe excesses. It is a commonly accepted that the excess of ^{136}Xe relative to the atmosphere results from Uranium fission where ^{136}I with a very short half-life (86 sec.) in ^{136}I$(\beta^-,\gamma)^{136}$Xe is the only ^{136}Xe precursor. The excesses of ^{130}Xe and ^{128}Xe can exclusively be explained by $\beta\beta$-decay of ^{130}Te and ^{128}Te respectively. However, to account for the excesses—relative to their atmospheric abundance—of the remaining isotopes, ^{129}Xe, ^{131}Xe, ^{132}Xe, and ^{134}Xe, presents a major problem. The possible origins of the ^{129}Xe and ^{131}Xe excesses might be different from that of ^{132}Xe and ^{134}Xe. On one hand, all four isotopes can be produced by the β-decay of Te, Sb and I precursors after their injection into the crystal lattice after mineral formation. Following Meshik (1988) and, Xe produced by this process is termed Chemical Fractionated Fission Xe (CFF-Xe). On the other hand, however, the excessive abundances of ^{129}Xe and ^{131}Xe could result from nuclear reactions following neutron-capture and/or resonance neutron capture by ^{128}Te and ^{130}Te, respectively (Browne and Berman, 1973): ^{128}Te$(n,\gamma)^{129}$Xe, ^{130}Te$(n,\gamma)^{131}$Xe, ^{130}Te$(\mu^-,n)^{129}$Xe ^{130}Te$(n,2n)^{129}$Xe.

We propose the introduction of a four-isotope diagram ^{131}Xe/^{134}Xe *vs* ^{129}Xe/^{132}Xe to distinguish the Xe-sources instead of three-isotope diagrams common in Xe geochemistry. Fig. 1 shows the positions of the various Xe sources. Some reservoirs have well accepted compositions: atmospheric Xe, CFF-Xe (Shukolyukov *et al.*, 1994), ^{235}U- and ^{238}U-fission Xe (cf. Table 1). The values for the isotopic compositions of CFF-Xe (Table 1) produced individually by the decay of (chemically) differently fractionated precursors were calculated with the formalism usually applied to the α-activity series of U and Th (Malyushev, 1981). To estimate the isotopic compositions of the end-members, we postulate that some quantity of β-active Xe-precursors remain dissolved in water in the presence of Uranium (even at a low level) irrespective of the chemical composition of the rocks and physico-chemical parameters of the whole systems. The precursors (Sb, Te and I) could fractionate during crystallization because of their different geochemical properties. Consequently, the isotopic composition of Xe in a mineral would inherit the isotopic signature of the precursor. For example, we expect an isotopic composition close to CFF-Te in Au and Ag bearing telluride, and an isotopic composition close to CFF-

TABLE. 1. Isotopic composition of major Xe reservoirs, CFF-Xe, and of Xe calculated individually from its precursors and denoted CFF-I, CFF-Te, and CFF-Sb from I, Te, and Sb, respectively

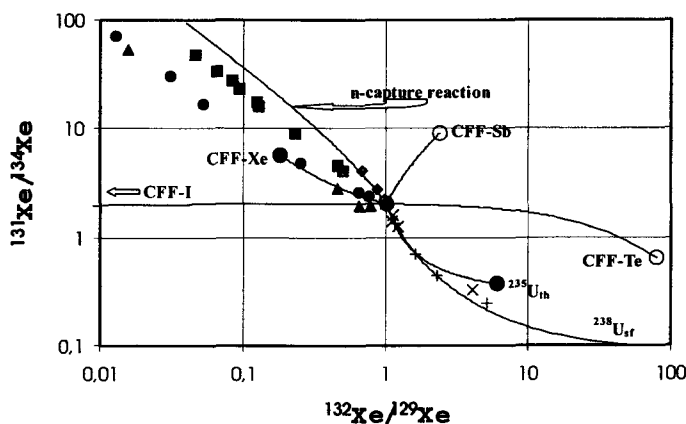| Ratio	CFF-Xe	CFF-I	CFF-Te	CFF-Sb	Atm.	^{238}U$_{sf}$	^{235}U$_{th}$
^{131}Xe/^{134}Xe	5.76	20.17	0.635	9.13	2.034	0.091	0.37
^{132}Xe/^{129}Xe	0.18	0.0001	79.66	2.40	1.017	297.5	6.04

FIG. 1. Four-isotope diagram $^{131}Xe/^{134}Xe$ vs. $^{129}Xe/^{132}Xe$ of CFF-Xe reservoirs and Xe isotopic data of tellurides and selenides. The rhombs and squares mark the compositions of Cripple Creek calaverite and Vulcan Mine tellurium, respectively (Bernatowicz et al., 1993); the triangles and full circles both represent tellurobismutite from Boliden (Srinivasan et al., 1972; M.M. Fugzan, priv. comm.). Crosses are new data from berzelianite and klaustalite selenides from Uranium deposits.

Sb in Sb-bearing sulphides. It should be noted that the time to reach steady state is short and comparable to the half-lives of Xe precursors. The only exception is ^{129}I with 17 Ma half-life. The production rate of a precursor isotope can be calculated from (with an Iodine isotope as an example):

$$\frac{d[^iI]}{dt} = \frac{[U] \times \Sigma y(^iI)}{T_{1/2}(U)} - \frac{[^iI]}{T_{1/2}(^iI)} \quad (1)$$

where t is time in years, $\Sigma y(^iI)$ is the cumulative yield of the ^iI isotope in β-decay series, $T_{1/2}$ is the half-life of the radioactive isotope, square brackets denote concentrations.

We have assumed that the cumulative yield of the precursor isotope is equal to the cumulative yield of the corresponding Xe isotope

At steady state $d[^iI]/dt = 0$. Substituting this to Eqn. (1), we obtain for the ratio of two isotopes

$$\frac{[^iI]}{[^jI]} = \frac{T_{1/2}(^iI) \times \Sigma y(^iI)}{T_{1/2}(^jI) \times \Sigma y(^jI)} \quad (2)$$

Certainly the isotopes of Iodine can be replaced with isotopes of any other precursor element.

The Xe isotopic compositions calculated with Eqn. (2) from the isotopic compositions of the precursors are listed in the Table 1 and are shown in Fig. 1. There the line of Xe production from epithermal n-capture (n,(-reactions on ^{128}Te and ^{130}Te) is calculated by mixing in different proportions atmospheric Xe and the calculated two-isotope gas that has a ratio $^{129}Xe/^{131}Xe = 0.68$.

Advantages of the newly introduced four-isotope diagram are:

i) All Xe isotopes involved in CFF-Xe formation and from neutron capture processes are presented simultaneously.

ii) Xe isotopic compositions resulting from different sources are presented in this diagram by points. This allows a more reliable interpretation of experimental data as in three-isotope diagrams.

iii) This diagram allows to study important Xe isotope geochemical processes like contamination and mixing of Xe components from different reservoirs (Xe from the mantle, the atmosphere, from neutron-induced U fission and from spontaneous fission of U products, and of CFF-Xe).

To demonstrate the usefulness of this new approach we apply it to investigate the origins of CFF-Xe. CFF-Xe was determined by Shukolyukov et al. (1994) and its composition is in between the (n,γ)-line and the atmosphere-CFF-I mixing line (Fig. 1). From this position we conclude that CFF-Xe was formed simultaneously by both processes.

References

Shukolyukov, Yu.A, Jessberger, E.K., Meshik, A.P., Minh Dang, Vu, Jordan, J.L. (1994) Geochim. Cosmochim. Acta, **58**, 3075–17.

Meshik, A.P. (1988) Ph. D. Thesis, 213p., Vernadsky Institute, Moscow, (in Russian).

Browne, J.C. and Berman, B.L. (1973) Phys.Rev., **C8**, 2405–6.