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Thorium and uranium abundances in the Jilin H5 chondrite

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Thorium and uranium abundances have been measured in the Jilin H5 chondrite by radiochemical neutron activation analysis. Although the abundances of (35.8 ± 1.5) ppb Th and (14.9 ± 0.9) ppb U are within the range of literature values, the ratio Th/U is about 25% lower than the average value for H chondrites. Terrestrial addition of uranium appears to be the most likely explanation.

1. Introduction

Begemann et al. [1] calculated U/Th-^{4}He ages for three samples of the Jilin H5 chondrite using average H chondrite concentrations of Th and U [2] and obtained different ages for different samples. Similar differences were observed by the same authors for the ^{40}K-^{40}Ar ages, which suggests that some fractions of ^{4}He and ^{40}Ar were lost by diffusion. Although the error introduced by assuming average chondrite values for Th and U is probably small, it seemed nevertheless desirable to determine their actual concentrations to check the possibility that inhomogeneous distribution of Th and U could have contributed to the observed differences in the U/Th-^{4}He ages.

2. Method

Most results for Th and U in meteorites have been obtained by neutron activation analysis [2] because of its sensitivity and freedom from problems of reagent contamination in chemical processing of the sample. Therefore we adopted the general approach developed by Bate et al. [3] and Morgan and Lovering [4] with major revisions in the chemical separation scheme.

Samples of about 1 g were carefully crushed in an agate mortar and the metallic fraction was separated with a hand magnet. Duplicate aliquots of about 100 mg of silicate fractions were sealed together with U and Th standard in pure silica tubes and irradiated for 24 hours at a flux of 1×10^{14} n/cm^2 s in the FR2 reactor of the Kernforschungszentrum Karlsruhe.

For the determination of Th and U concentrations, reactions

$$^{232}\text{Th}(n, \gamma)^{233}\text{Th} \xrightarrow{\beta^-} {}^{233}\text{Pa}$$

and

$$^{238}\text{U}(n, \gamma)^{239}\text{U} \xrightarrow{\beta^-} {}^{239}\text{Np}$$

were utilized. This necessitated the separation of ^{233}Pa and ^{239}Np from the matrix activity, for which radiochemical separation schemes have been worked out using ^{231}Pa and ^{237}Np as tracers [3,4]. Parts of these schemes have been used in this work, but the advance of high-resolution gamma detector technology allowed several simplifications, because the requirements for radiochemical purity of samples for measurement are not as stringent any more. Small amounts of radioactive impurities from the matrix can be resolved and do

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not disturb the measurements of ^{233}Pa and ^{239}Np. In addition, instead of using alpha spectrometry to determine the chemical yield we reirradiated aliquots of the sample solution for 4 hours at a flux of 1×10^{13} n/cm^2 s and measured the gamma rays of ^{232}Pa ($t_{1/2} = 1.32$ d) and ^{238}Np ($t_{1/2} = 2.12$ d), which were produced by (n, γ) reactions from the tracers.

3. Analytical techniques

Preparation of standards: Stock solutions were prepared from analytical grade thorium nitrate and uranyl nitrate. Immediately before use, appropriately diluted solutions were prepared, from which aliquots were pipetted onto a clean aluminium foil.

Preparation of tracers: A stock solution ^{231}Pa was prepared by dissolving 0.1 mg ^{231}Pa as oxide in 10 ml of 6 M HCl/1 M HF. Immediately before use, this stock solution was appropriately diluted. The ^{237}Np stock solution contained 1 mg of ^{237}Np as oxide and 6 M HCl. Immediately before use, an appropriate amount of this stock solution was purified on an anion-exchange column as described in the radiochemical procedure and the time of elution noted.

After irradiation, the samples were transferred to platinum crucibles and 1.5 μg of ^{231}Pa and 5 μg of ^{237}Np were added in the form of dilute acid solutions. In a polyethylene flask, the same amounts of tracer were diluted to the final volume of the sample solution after chemical separation (3 ml) and two aliquots of 50 μl were pipetted into small polyethylene vials and dried in a desiccator. These aliquots served as 100% comparators for the determination of the chemical yield. The samples were decomposed with an HF-$HClO_4$ mixture, taken to dryness and digested in 6 M HCl until a clear solution was obtained. Concentrated HCl was added to make the solution 9 M in HCl, which was then transferred to a Dowex 1×8 anion exchange column. After washing with three column volumes of 9 M HCl, Np was eluted with 4 M HCl. At this stage Np was still contaminated with ^{60}Co so that a further purification step was necessary. Pa was removed from the column with 6 M HCl/0.5 M HF, the eluate taken almost to dryness, diluted with 6 M HNO_3 to 3 ml and transferred to a counting tube.

The Np fraction was prepared for extraction with TTA/Xylol from a solution containing 0.5 M hydroxylamine, 0.125 M ferrous chloride and 1 M HCl. After back extraction with 10 M HNO_3, the nitric acid was fumed off with HCl and the solution containing 9 M HCl was again loaded on a Dowex 1×8 anion exchange column. Np was eluted with 4 M HCl, brought to a volume of 3 ml and transferred to a counting tube. Aliquots of 50 μl were pipetted from the Pa and Np counting solutions into small polyethylene vials, dried in a desiccator and reirradiated together with the comparators to determine the chemical yields, which were typically 90% for Pa and 60% for Np. The standards were treated in a similar way except that the extraction and the second anion exchange step were omitted for Np. All counting was performed on a 64 cm^3 Ge(Li) well-type detector with a resolution of about 2.7 keV at 1.33 MeV. The results for Pa had to be corrected for the contribution by the ingrowth of ^{233}Pa from the ^{237}Np tracer. At the beginning of this study no ^{231}Pa tracer was available so that Th values were obtained without determination of the chemical yield. These values are given in brackets in Table 2. In a second run only Th was analyzed, so that a correction for ^{233}Pa from ^{237}Np was not necessary.

Finally, the results for the silicate fraction were used to calculate whole rock concentrations assuming negligible Th and U contents in the metal phase. This appears to be justified, because the few analyses of these elements in meteoritic metal, which do exist, indicate concentrations in the order of 10^{-11} g/g [2]. Instrumental determination of Sc in both silicate and magnetic fraction showed that the magnetic fraction contained about 10% silicate, which was taken into account.

4. Results and discussion

For comparison, samples of the U.S. Geological Survey standard BCR-1 (Columbia River basalt) [5] and of an Allende meteorite reference sample [6] were analyzed. The results are listed in Table 1

TABLE 1

Thorium and uranium abundances in BCR-1 and Allende

Sample	Th (μg/g)	Method [a]	U (μg/g)	Method [a]	Reference
BCR-1	6.0 (r) [b]	various	1.74 (a) [b]	various	[5]
	5.8	NAA	1.68	NAA	[7]
	6.24 [c]	NAA	1.83 ± 0.10	NAA	this work
	6.36 ± 0.25	NAA	–	NAA	this work
Allende reference sample	0.070	NAA	–		[6]
	0.063	SSMS	–		[6]
	–		0.016	NAA	[6]
	–		0.0163 ± 0.004	NAA	[13]
	0.049 ± 0.004	NAA	0.0152 ± 0.0014	NAA	this work
Allende	0.0622	MS	0.0153	MS	[8]
	–		0.0143	MS	[9]
	0.056	SSMS	0.017	SSMS	[10]
	–		0.0165	FT	[11]
	–		0.0155	FT	[11]
	–		0.0162	NAA	[12]

[a] NAA = neutron activation analysis, MS = mass spectrometry with isotope dilution, SSMS = spark source mass spectrometry, FT = fission tracks.

[b] Values are qualified in Flanagan [5] in decreasing order as recommended (r), average (a) and magnitude (m).

[c] Without determination of the chemical yield.

and show good agreement for U with both reference samples. Our Th value for Allende is somewhat lower than those obtained by other workers, but since our Th value for BCR-1 is slightly higher than the recommended value no systematic error is indicated.

The results for Jilin (Table 2) show that both elements are homogeneously distributed over the whole mass and that the observed differences in the U/Th-^{4}He ages are only due to different diffusion losses of ^{4}He.

TABLE 2

Thorium and uranium abundances in Jilin. Values in parentheses have been obtained without determination of the chemical yield

Sample	Th (ng/g)	U (ng/g)	Th/U
Jilin 1-2	(29.5)	15.5 ± 1.7	
	38.6 ± 3.1 [a]	–	2.4
Jilin 1-4	(36)	15.3 ± 2.0	
	35.6 ± 2.5	–	2.3
Jilin VI-42-04	(34)	14.7 ± 0.8	
	34.3 ± 2.4	–	2.3

[a] Errors were estimated from duplicate samples for U and from counting statistics for Th.

Compared to average abundances in H-chondrites [1] our results for Jilin are somewhat higher in U and slightly lower in Th. Consequently, the Th/U ratio is about 25% lower than the average value for H chondrites [14,15]. A geochemical fractionation of the two refractory lithophile elements seems unlikely. Contamination of the samples used for analysis during transport or laboratory preparation is also unlikely because of the excellent agreement of Th and U values in three samples from separate meteorite fragments. From such a type of contamination one would expect much larger scatter of the data. There remains the possibility of contamination during the deposition time in the ground. Although the meteorite fragments were recovered already about a month after the fall, they are reported to have been exposed to groundwater, which led to rust stains [1]. Since U is much easier mobilized by groundwater than Th, contamination with groundwater would result in low Th/U ratios in the meteorite.

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