

URANIUM ISOTOPIC ABUNDANCE IN ALLENDE RESIDUE

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Recently the results from isotopic analyses of various elements extracted from etch fractions of an acid resistant residue from Allende were discussed [1,2]. Large isotopic enrichments were found in ^{142}Nd and ^{143}Nd (0.47% and 36%) which were attributed to the alpha decay of now extinct ^{146}Sm and long lived ^{147}Sm . It was concluded that the isotopic enrichments were greatly magnified by recoil of the product nuclei into a carbon layer coating rare earth bearing grains. If this mechanism is indeed responsible for the observed Nd effects it should also manifest itself in the uranium isotopic system. In particular, the alpha decay product of ^{238}U is ^{234}U which then should also be enriched in the same fractions where the Nd effects are found.

We analyzed the U isotopic composition in two etch fractions from the Allende acid resistant residue (Allende CD), CF-1 (gas poor carbon) and CG-1 (gas rich carbon and chromite) [1]. For the present experiment, the starting materials were the solutions remaining from the previous study [1,2]. After addition of weighed amounts of combined $^{233,236}\text{U}$ spike the sample solutions were passed through the usual two anion columns to facilitate clean separation of U from other elements [3]. Since the amounts of U available for measurement were expected to be extremely small (<1 ng) an attempt was made to improve sample loading procedures for mass spectrometry. Fine high purity Re powder was placed into a canoe shaped Re filament and sintered at high temperature during the final stage of filament bakeout. The sample solution was loaded onto the Re powder and was covered with high purity fine grained graphite after the sample was dried. Stable ion beams of 10^{-13} A were obtained for several hours for 1 ng samples. However, we were required to keep ion beam intensities for ^{238}U below 5×10^{-14} A due to nonlinearities at higher beam intensities caused by the recently replaced photomultiplier. At the lower signal levels 10 standard runs showed good internal reproducibility and agreed with our earlier measurements [3] of the 235/238 ratio to within better than 0.1%.

The results for NBS standard U-960 obtained under the above conditions on 0.4 to 1 ng of U are given in Table 1. For two of the ten standard runs 234/238 was also measured. The result of $^{234}\text{U}/^{238}\text{U} = 0.000055$ is in excellent agreement with the normal value with an external precision of better than 10% for sample sizes of ~ 1 ng. For samples CF-1 and CG-1 the calculated amounts were much smaller (66 and 89 pg) and represent lower limits of the actual U present in the original samples because the double spike was added to already processed solutions; we estimate, however, that at least 80% of the total U was recovered. The most striking result is the high enrichment in ^{234}U in both samples. Especially for CF-1 the 234/238 ratio is 26x higher than the normal equilibrated value and is far above any possible systematic error which might be caused by the detector system. We take these enrichments as being due to recoil following alpha decay of ^{238}U . These results are entirely consistent with and confirm our earlier conclusion for the Sm-Nd system [2].

The ratio of excess atoms of ^{234}U in CF-1/CG-1 is 1.8. The same value is obtained for excess ^{143}Nd in the same etch fractions [2]. Note that the recoil energy of ^{234}U is only slightly higher (15%) than that of ^{143}Nd (from ^{147}Sm) and basically the same as ^{142}Nd (from ^{146}Sm). Hence, the identical distribution of recoiled U and Nd atoms indicates that the sighting of their parent U and Sm atoms in the associated mineral grains also coincide. Because of the large difference in half lives of ^{234}U (2.4×10^5 yr) and ^{147}Sm (1.06×10^{11} yr) we can further conclude that the configuration within the meteorite of the acid resistant phases was stable over the age of the meteorite. As a consequence of the short half life, the enrichment in ^{234}U directly reflects the lower limit of the U concentration in the mineral grains adjacent to or covered by the acid resistant carbon layer as well as the trapping efficiency and/or geometric configuration of the various components of the residue. The excess ^{234}U atoms we measured are 1×10^6 for CF-1 and 5.4×10^5 for CG-1. Expressed as U concentration reflected in the particular etch fraction we obtain 17.6 ppb for CF-1 and 37 ppb for CG-1. For an ideal case, where the total layer thickness or the radius of a layered carbon grain is comparable to the recoil range, the actual U concentration in the adjacent mineral grains is at least twice as large. For less ideal configurations it will be even higher. Also, some of the recoiled U may have been lost during the preparation of the residue. These U contents are much higher than typical bulk values for Allende (~ 15 ppb, [4]) and suggest that

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at least part of the coated mineral grains consists of phases enriched in refractory elements. A similar conclusion can be reached by using the published excess ^{143}Nd data for the same samples [2]. We further note that CG-1 contains more than twice the number of recoiled U atoms per weight of sample than does CF-1. This indicates a much higher trapping efficiency for CG-1 which may be due to its chemical components or its geometrical configuration relative to CF-1 material within the meteorite (e.g., layered carbon grains?). But it could simply indicate the higher resistance of CG-1 during chemical treatment.

Also presented in Table 1 are the results for ^{235}U . The 235/238 ratios of CF-1 and CG-1 are both slightly below our normal. In order to search for a possible instrumental bias or small isobaric interference which may only become significant at very low signal levels we measured a comparably small standard sample. As is apparent from the result (Table 1), the 235/238 ratio is also low while the 234/238 ratio agrees with our normal value within error. It is clear that the lower than normal 235/238 values are an artifact. The reason for this is not understood but we consider the most likely cause an overestimation of the decay time constant of the photomultiplier.

References: [1] Lewis *et al.* (1983), *Science* 222, 1013-1015. [2] Lugmair *et al.* (1983), *Science* 222, 1015-1018. [3] Shimamura and Lugmair (1981), *LPSC XII*, 976-978. [4] Tatsumoto *et al.* (1973), *Science* 180, 1279-1283.

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Table 1: Uranium in etch fractions from Allende residue CD

Sample	U [ng]	235/238	234/238	[U] ppb	^{234}U exc. atoms
U-960	0.4-1.0	0.007254 ^{a)}	0.000055 ^{b)}		
		5	4		
CF-1	0.066	0.00720	0.001426	0.70	2.3×10^8
		3	23		
CG-1	0.089	0.00722	0.00061	3.66	1.2×10^8
		4	5		
U-960 Small	0.1	0.00712	0.000063		
		6	17		

a) average of 10 sample runs.

b) average of 2 runs with maximum probable error.