

THE CASE FOR AN UNFRACTIONATED $^{244}\text{Pu}/^{238}\text{U}$ RATIO IN HIGH-TEMPERATURE CONDENSATES

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The coarse-grained, Ca-rich inclusions in the Allende meteorite are the highest-temperature condensates from the cooling solar nebula and, as such, the oldest solid objects in the solar system. All refractory elements with condensation points above the accretion temperature of the inclusions whose concentrations in them have been measured are seen to be present in the inclusions in unfractionated proportion to one another relative to C1 chondrites when data are averaged for a large number of inclusions. Observational data for U and theoretical data for both U and Pu suggest that these elements exhibited refractory behavior in the solar nebula. An experiment is proposed in which fissionogenic Xe and U contents are measured in a suite of these inclusions to obtain the $^{244}\text{Pu}/^{238}\text{U}$ ratio of the solar system at the time of initial condensation with an uncertainty of $\pm 15\%$.

1. Introduction

^{244}Pu ($t_{1/2} = 83 \times 10^6$ yr) is known to be an extinct radionuclide whose decay products are present in meteorites [1,2]. Its usefulness as a chronometer has been hampered, however, mainly because its abundance must be related to that of a long-lived isotope of the nearest actinide, U, since no stable isotope of Pu exists. Therefore, an exact knowledge of the $^{244}\text{Pu}/^{238}\text{U}$ ratio at the time of initial condensation of the solar system is necessary in order to better define the length of time between that event and cessation of r-process nucleosynthesis and to calculate time intervals between condensation and formation of different objects in the solar system. The problem is that radioactive decay is not the only process which causes variation in the $^{244}\text{Pu}/$

^{238}U ratio. Because the chemical properties of Pu are slightly different from those of U, large changes in the $^{244}\text{Pu}/^{238}\text{U}$ ratio might have occurred during the processes leading to formation of meteorite parent bodies and planets. Gray et al. [3] commented, "If there existed 'high temperature' phases which were very retentive for Xe and which also contained Pu and U or Th in unfractionated proportions, then it would be possible to utilize this approach... However, no evidence has yet been presented which would support so idealized a scheme". In this communication, we wish to demonstrate that chemical properties of the coarse-grained, Ca-rich inclusions in the Allende meteorite suggest that they are the ideal materials longed for by Gray et al. [3] in which the initial $^{244}\text{Pu}/^{238}\text{U}$ ratio at the time of condensation can be measured.

2. Discussion

2.1. Origin of Allende inclusions

The coarse-grained, Ca–Al-rich inclusions in the Allende meteorite are thought to be the earliest, high-temperature condensates from the cooling vapor of the solar nebula [4,5], representing ~5 wt.% of the total condensable matter [6]. These inclusions are several millimeters to 2 cm in size, subspherical to angular in shape and white in color, sometimes with gray patches. They are hard, dense objects which are resistant to scraping and crushing. Using thermodynamic calculations, Grossman [7] demonstrated that the mineralogy of some of them, melilite + spinel + perovskite ± diopside, is consistent with their condensation at temperatures $>1450^{\circ}\text{K}$ at 10^{-3} atmosphere total nebular pressure, before crystallization of significant fractions of Fe, Mg and Si which constitute the bulk of the condensable matter of the solar system. Thus, theoretical considerations suggest that they are the oldest crystalline materials in the solar system.

2.2. Age of Allende inclusions

Radiometric age determinations [3,8,9] support these earlier suggestions that the high-temperature condensate inclusions are extremely ancient. In particular, Gray et al. [3] found one inclusion having the lowest $^{87}\text{Sr}/^{86}\text{Sr}$ ratio ever seen in any solar system object. In addition, the extreme antiquity of these inclusions is further demonstrated by the presence in them of excess ^{26}Mg attributed to in-situ decay of 7.4×10^5 yr ^{26}Al [10,11].

2.3. Composition of Allende inclusions

Grossman [6] showed that many trace elements are so refractory or form such stable oxides that they should have condensed in the same high-temperature range as the major mineral phases of the coarse-grained inclusions. Many trace element studies [6,12–21] have demonstrated enrichment of the inclusions in these refractory elements and their depletion in elements which are more volatile in the solar nebula. The concentration of any particular refractory trace element and, often, the ratio of one refractory to another vary widely from inclusion to inclusion. This suggests

that the refractories condensed as grains of several different phases and that different proportions of these phases were incorporated by different inclusions. Grossman and Ganapathy [18,20] and Ganapathy and Grossman [21] pointed out that an amazing regularity is preserved in the compositions of coarse-grained inclusions in spite of this variability. They divided the mean concentration of each refractory element in a suite of nine inclusions by its abundance in C1 chondrites and found, for each of the 20 refractories they determined, that this ratio is remarkably constant at 17.7 ± 1.8 (see Table 1). Because C1 chondrites are thought to have the mean composition of the total condensable matter of the solar system, the uniformity of this ratio shows that *the refractory elements are not fractionated significantly from one another* in a sample consisting of a large number of inclusions. Fig. 1

TABLE 1

Refractory elements * in a suite of nine coarse-grained inclusions from the Allende meteorite (values in ppm unless otherwise indicated)

Element	Mean concentration	Standard deviation (%)	Enrichment factor (relative to C1 chondrites)
Ca	17.2%	32.1	17.7
Sr	129	30.4	15.1
Ba	39.7	43.3	16.5
Sc	118	60.3	18.5
La	4.24	30.4	16.8
Ce	13.7	48.6	21.3
Sm	2.80	32.9	18.2
Eu	1.13	19.5	19.3
Tb	0.60	58.3	15.4
Dy	4.90	49.8	19.4
Yb	2.68	38.1	16.0
Zr	59.4	58.2	19.2
Hf	1.84	66.3	18.4
Ta	0.24	45.8	17.9
W	1.88	56.9	21.4
U	163 ppb	39.2	18.0
Re	586 ppb	45.2	16.6
Ru	10.5	47.1	15.3
Os	8.11	46.7	16.9
Ir	8.16	49.9	15.9
Mean 17.7 ± 1.8			

* Sources of data: INAA measurements in [18] and RNAA measurements in [20,21], excluding inclusion 5.

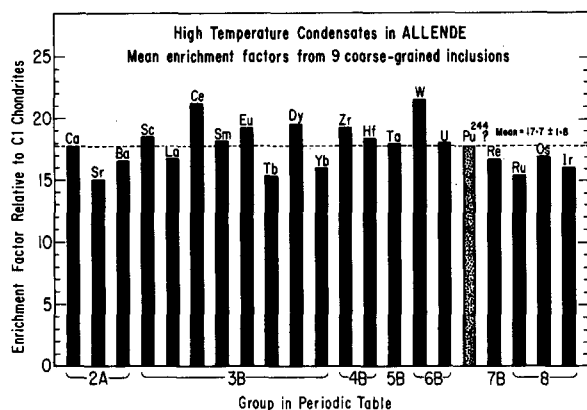


Fig. 1. Elements having condensation temperatures greater than the accretion temperature of the coarse-grained inclusions (1450°K at 10^{-3} atmosphere total nebular pressure) are all enriched in them relative to their abundances in C1 chondrites. The enrichment factors are all nearly the same, however, indicating that the refractory elements have not fractionated significantly from one another relative to their solar system abundances.

shows that the elements investigated come from all groups in the periodic table and possess, therefore, drastic differences in geochemical behavior from one another. The only property in common to all of these elements is that they have condensation temperatures from a gas of solar composition which are higher than the accretion temperature of the inclusions, 1450°K . Theoretical calculations [6] suggest, although these elements began to condense at widely different temperatures from one another in the range $1500\text{--}2000^{\circ}\text{K}$, that each one should have been virtually totally condensed at 1450°K . The enrichment factor of 17.7 relative to C1 chondrites indicates that the refractories were efficiently incorporated into $100/17.7 = 5.6$ wt. %

of the total condensable matter, a value in agreement with condensation calculations [6,7]. Thus, while individual inclusions are not typical of the mean composition of the high-temperature condensates, a collection of inclusions, being a larger sample of this material, is far more representative.

2.4. Condensation of Pu, U and Th

In order to calculate the condensation temperatures of Pu, U and Th, we have assumed that the important gaseous molecules and crystalline phases are the elements and their oxides. All species included in the calculation are listed in Table 2. Thermodynamic data were taken from the compilation of Ackermann and Chandrasekharaiah [22] except for values for U_3O_8 [23] and gaseous monatomic O [24]. P_0 as a function of temperature was taken from programs used to compute the condensation sequence at 10^{-3} atmosphere total pressure in Grossman [25]. Abundances of U and Th were taken from Cameron [26] and Ganapathy and Anders [27], respectively. The Pu abundance in the early solar system was calculated by assuming that ^{244}Pu was the only isotope and that the initial $^{244}\text{Pu}/^{238}\text{U}$ ratio was 0.015 [28]. The method of calculation is the same as that outlined in [6].

ThO_2 , having a condensation point of 1555°K , is the only phase listed in Table 2 which reaches saturation in the solar nebula above 1450°K at 10^{-3} atmosphere total pressure. None of the U- or Pu-bearing crystalline phases in Table 2 can do so. The first condensates of these elements, UO_2 and PuO_2 , would not have condensed as pure phases until the temperature fell to $\sim 1300^{\circ}\text{K}$ and $\sim 1200^{\circ}\text{K}$, respectively. As seen in Fig. 1, however, U is normally enriched in the inclusions, indicating that it must have condensed in some

TABLE 2

Elemental and oxide species of Th, U and Pu for which free energies of formation are available

Element	Gaseous molecules	Crystalline phases	Condensation temperature of pure phase ($^{\circ}\text{K}$)
Th	Th, ThO, ThO ₂	Th, ThO ₂	ThO ₂ : 1555
U	U, UO, UO ₂ , UO ₃	U, UO, U ₂ O ₃ , UO ₂ , U ₃ O ₈	UO ₂ : ~ 1300
Pu	Pu, PuO, PuO ₂	Pu, PuO, Pu ₂ O ₃ , PuO ₂	PuO ₂ : ~ 1200

form above 1450°K. This is analogous to the case of the rare earth elements (REE) which are all normally enriched in the inclusions despite the fact that some of their pure oxides also do not reach saturation at these temperatures [18]. In order to explain this phenomenon, Grossman and Ganapathy [18] suggested that REE may have condensed as one or more complex refractory phases for which no thermodynamic data are available. This could have happened to U and Pu also. Indeed, such condensate minerals may have already been discovered as Lovering et al. [29] have reported several trace U-, Th-, REE-rich phases in coarse-grained inclusions in Allende.

Another explanation proposed for REE enrichment in the inclusions [6,18] is that they condensed in solid solution in previously formed phases, such as perovskite, which contain elements of comparable or greater abundance. Applying this idea to the cases of U and Pu, we note that a terrestrial perovskite is known which contains 360 ppm U [30], indicating a tolerance of its crystal structure for this element. Because Pu and U have such similar ionic radii, Pu is also expected to enter readily into the perovskite lattice. PuO_2 and UO_2 , respectively, have the highest activities at 1500°K in the solar nebula of all the Pu- and U-bearing crystalline phases in Table 2. Assuming ideal solid solution and ionic behavior, we calculate from the above thermodynamic data that perovskite would contain 0.2 ppm Pu and 16 ppm U at this temperature. Because virtually all of the Ti is condensed as perovskite at 1500°K and because Ti is so abundant, these concentrations represent 98% condensation of Pu and 99% condensation of U. These per cents are independent of the cosmic abundances of Pu and U.

Theoretical calculations thus show, as in the case the REE, that Pu and U would have exhibited refractory behavior in the solar nebula even if they did not condense as pure compounds. By analogy to all the other refractories, they are expected to be present in the coarse-grained inclusions in unfractionated proportion to one another.

2.5 Proposed experiment to measure the initial $^{244}\text{Pu}/^{238}\text{U}$ ratio

From the above discussion, the refractory elements Pu and U are expected to have been incorporated into the coarse-grained inclusions in Allende during the high-temperature condensation stage of the solar nebula.

On the average, each of these elements should have reached a concentration level corresponding to 17.7 times its abundance in C1 chondrites, resulting in a $^{244}\text{Pu}/^{238}\text{U}$ ratio identical to that of the solar nebula, assuming that the Pu- and U-bearing grains condensed in our solar system. Larimer and Anders [31] and Clark et al. [32] showed that the cooling time of the solar nebula, and presumably condensate particles immersed in it, was less than 10^6 years. Thus, it is likely that the inclusions began retention of fissiogenic Xe from decay of ^{244}Pu within an interval of time after the beginning of condensation which was extremely short relative to the half-life of ^{244}Pu . Further, although later metamorphism has disturbed the Rb–Sr [3] and U–Pb [9] isotopic systems in different types of inclusions in Allende, many mineralogical, textural, chemical and isotopic features suggest that this event was not caused by intense heating. Thus, loss of fission Xe during metamorphism is likely to have been negligible, particularly since it is rather tightly held in chondrites [33]. Because the inclusions are high-temperature condensates, the correction for the amount of primordial trapped gas is expected to be very small. This is in sharp contrast to C1 chondrites which, theoretically, should also possess an unfractionated $^{244}\text{Pu}/^{238}\text{U}$ ratio but which, in practice, contain so much trapped gas that the fission component cannot be seen.

There also exists in Allende a class of objects, fine-grained inclusions, in which refractory elements are severally fractionated from one another, both in individual inclusions [13] and in groups of inclusions as a whole [14,19]. Although these are easily distinguished on the basis of their pink or purple color and soft, sugary consistency from coarse-grained ones, Grossman and Ganapathy [19] discovered one inclusion among their group of ten so-called coarse-grained inclusions which had all the physical attributes of a coarse-grained one but the characteristic, peculiar fractionation pattern of a fine-grained one. They suggested that it was formed by melting and recrystallization of a fine-grained inclusion. Chemically, one of the easiest and most straightforward ways of distinguishing coarse-grained inclusions from fine-grained ones, whether re-melted or not, is by their Ir contents. Coarse-grained inclusions usually contain 6–12 ppm Ir with values sometimes as low as 0.6 ppm. Fine-grained inclusions usually contain several ppb Ir with occasional values as high as 300–400 ppb.

We propose the following experiment to measure the value of the $^{244}\text{Pu}/^{238}\text{U}$ ratio of the solar system at the time of initial condensation. Collect at least 10 mg of each of ten randomly-selected coarse-grained, Ca-rich inclusions from the Allende meteorite. Measure the Ir content of each inclusion. If they all contain >0.5 ppm Ir, mix all samples together and measure the bulk U content and abundance of each Xe isotope in the total sample. If any inclusion contains <0.5 ppm Ir, reject it as a re-melted fine-grained inclusion and analyze the bulk sample consisting of only the remaining inclusions. Calculate the $^{244}\text{Pu}/^{238}\text{U}$ ratio from the fissiogenic Xe content and U abundance. It is not necessary to measure the U and Xe content of each inclusion individually unless information is needed about the variability of this ratio from inclusion to inclusion which, in turn, has implications for the nature of the host condensate phases.

2.6. Existing information on the initial $^{244}\text{Pu}/^{238}\text{U}$ ratio

The $^{244}\text{Pu}/^{238}\text{U}$ ratio inferred for the bulk LL6 chondrite St. Séverin, 0.015 [28] is currently the best estimate of the value characteristic of the early solar system. Use of this ratio in calculating the length of time between cessation of r-process nucleosynthesis and cooling of meteoritic materials to below Xe retention temperatures rests critically on the assumption that Pu and U did not fractionate from one another relative to their solar system abundances during the formation of St. Séverin. In this regard, it is very troubling that the inferred initial $^{244}\text{Pu}/^{238}\text{U}$ ratio for whitlockite separated from St. Séverin is 0.033 [34]. Although whitlockite accounts for only a small fraction of the total U in the chondrite [35], the fact that its ratio is a factor of 2.4 higher than that for the bulk meteorite is a warning signal that large-scale fractionation of Pu from U took place during formation of the components of St. Séverin. There is no reason to assume that all Pu- and U-bearing complementary components accreted into the meteorite in exactly the same proportions in which they formed. A further problem is that results of partitioning experiments of Pu and U [36,37] indicate that relatively minor ($\sim 20\%$) differences are expected in the $^{244}\text{Pu}/^{238}\text{U}$ ratios of coexisting whitlockite and clinopyroxene. To the extent that these data can be applied to actual conditions of meteorite formation

and actual meteoritic materials, they suggest that St. Séverin whitlockite did not equilibrate its $^{244}\text{Pu}/^{238}\text{U}$ ratio with the other components of the meteorite either before or after its assembly. The possibility exists, therefore, that the whitlockite formed in equilibrium with matter having a $^{244}\text{Pu}/^{238}\text{U}$ ratio of ~ 0.03 but was later added accidentally to material from another system with a lower $^{244}\text{Pu}/^{238}\text{U}$ ratio to form St. Séverin. The advantage of the experiment proposed in this paper is that there are both theoretical and experimental data which predict that the measured $^{244}\text{Pu}/^{238}\text{U}$ ratio will be within 15% of the solar nebular value.

Has this experiment not already been performed? Podosek and Lewis [38] measured the fission Xe and U contents of a sample consisting of several fragments of "white inclusions" handpicked from a crushed specimen of Allende. If these were truly coarse-grained, high-temperature condensate inclusions, the inferred initial $^{244}\text{Pu}/^{238}\text{U}$ ratio of 0.087 calculated by them would imply either severe fractionation of Pu from U in St. Séverin or an earlier formation time for the inclusions by 2.2×10^8 yr relative to bulk St. Séverin. Unfortunately, however, the bulk U content measured in their sample is only 19 ppb. When this value is compared with that expected for coarse-grained inclusions in Table 1, 163 ppb, it must be concluded that high-temperature condensates were only a relatively minor component of the Podosek-Lewis "white inclusions" sample. Thus, neither of the above explanations for the high $^{244}\text{Pu}/^{238}\text{U}$ ratio is necessarily correct. It could have been caused by a large proportion of light-colored, fine-grained inclusions in their sample. These are known to have suffered severe chemical fractionations of some refractory elements relative to others [19], including heavy REE from light REE [13]. In the experiment proposed here, if fine-grained inclusions were accidentally sampled, they would be identified by their Ir contents and rejected from the sample before measurement of Xe and U contents.

There appears to be only one paper in the literature on bona fide coarse-grained inclusions from which some information bearing on the $^{244}\text{Pu}/^{238}\text{U}$ ratio is retrievable. Shirck [39] found a correlation between excess track density and U content in melilite crystals in a single inclusion from Allende and attributed the presence of excess tracks to fission of ^{244}Pu . Making use of the fact that the ratio of densities of excess tracks to ^{238}U spontaneous fission tracks is relatively

constant in Shirck's data ($1\sigma = 24\%$) and assuming equal track registration efficiencies for ^{244}Pu and ^{238}U , we calculate an initial $^{244}\text{Pu}/^{238}\text{U}$ ratio of 0.004. Because of the relative ease with which fission tracks can be annealed, this value should be taken as a lower limit to the true ratio for the melilite unless it is confirmed by Xe measurements. In any case, it cannot be assumed that the $^{244}\text{Pu}/^{238}\text{U}$ ratios of all other phases in the inclusion are the same as in the melilite. In fact, more recent data [40] show that pyroxene and melilite from the same inclusion have different ratios from one another. This suggests that different inclusions, containing different proportions of the component phases, have different bulk ratios. Need for the experiment proposed in this paper is again evident because it provides a means of averaging out refractory element fractionations seen in individual inclusions.

3. Conclusion

Lewis [41] wrote, "it is clear that any attempt at Pu/U dating will have to contend seriously with the possibility of chemical fractionations between plutonium and uranium in the samples under consideration. Any attempt to determine a standard initial $^{244}\text{Pu}/^{238}\text{U}$ ratio for the solar system should be based on a suite of carefully selected samples." In this paper, we have presented evidence that the coarse-grained, Ca-rich inclusions in the Allende meteorite represent such a suite. The experiment proposed herein should yield a value within 15% of the true $^{244}\text{Pu}/^{238}\text{U}$ ratio at the time of initial condensation of the solar system, thus eliminating a major stumbling block to further progress in Pu-Xe dating.

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