

PLUTONIUM-244 IN THE EARLY SOLAR SYSTEM?

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ABSTRACT

This paper presents results of noble gas, fission track, and petrographic investigations of inclusions in the Allende meteorite, most notably a single large coarse-grained "high temperature condensate" type inclusion. Discussion of the data focuses on the problem of whether or not ^{244}Pu actually was present in the early solar system, rather than being represented principally by fission xenon imported into the solar system in interstellar grains. The fission xenon content of the large inclusion corresponds to a $^{244}\text{Pu}/^{238}\text{U}$ ratio of 0.0161 ± 0.0019 at time of formation, 4.5×10^9 yr ago. Approximately 30% of this ^{244}Pu has produced visible fossil fission tracks in the major mineral phases; these decays must have occurred within the solar system, establishing a firm lower limit to the amount of ^{244}Pu which was present in the solar system. The actual value is almost certainly substantially greater than this lower limit.

Subject headings: abundances — meteors and meteorites

I. INTRODUCTION

Short-lived radionuclides figure prominently in theories of galactic nucleosynthesis and solar system evolution. Interest centers on species whose lifetimes are long enough to survive in detectable quantities from production in stellar nucleosynthesis to incorporation in solar system material, presently limiting the field to ^{129}I (half-life 17 million years), ^{244}Pu (83 million years), and ^{26}Al (0.7 million years). These nuclides are detected through their decay products in samples of high parent-to-daughter elemental ratios: ^{129}I through ^{129}Xe , ^{244}Pu through its spontaneous fission products ^{131}Xe , ^{132}Xe , ^{134}Xe and ^{136}Xe , and ^{26}Al through ^{26}Mg . Plutonium-244 is also detectable through tracks (trails of lattice disorder in dielectrics) produced by its fission fragments. Identification of these radionuclides in natural samples is limited to the most ancient accessible samples of the solar system, mainly meteorites. Conclusive identification of ^{129}I has been made through correlation of ^{129}Xe with ^{128}Xe artificially produced by neutron irradiation of ^{127}I (Jeffery and Reynolds 1961). Similarly conclusive identification of ^{244}Pu has been made by observation of the match between fission xenon spectra in meteorites and in artificially produced ^{244}Pu (Alexander *et al.* 1971). The case for ^{26}Al (Gray and Compston 1974; Lee, Papanastassiou, and Wasserburg 1976) is based on correlation of ^{26}Mg with ^{27}Al .

One view of solar system history stipulates that the dust and gas from which the Sun and at least the terrestrial planets (including meteorites) were eventually made passed through a phase of temperatures high enough to vaporize all preexisting solids. This would result in isotopic homogenization, in agreement with the generally observed isotopic uniformity of terrestrial, lunar, meteoritic, and, to the extent

measurable, solar materials. In this picture, the many well-known isotopic irregularities attributed to mass-dependent fractionation processes and nuclear reactions have been generated after the initial homogenization. In particular, the isotopic effects attributed to decay of ^{129}I and ^{244}Pu in meteoritic solid materials have been generated in situ, after the vaporization episode, requiring that ^{129}I and ^{244}Pu were present at the time of formation of the solar system.

In recent years, a number of meteoritic isotopic irregularities have been discovered which are not easily explained by the well-known mechanisms of isotopic fractionation, radioactive decay, and cosmic-ray-induced nuclear reactions. Most notable among these is enrichment of ^{16}O (Clayton, Grossman, and Mayeda 1973); other effects have been noted in neon (Black 1972), magnesium (Lee and Papanastassiou 1974), and xenon (e.g., Sabu and Manuel 1976; Pepin and Phinney 1976; Drozd and Podosek 1976). An attractive hypothesis for the ^{16}O enrichment is that the extra ^{16}O is carried in unhomogenized refractory interstellar dust which was not vaporized in the early stages of solar system evolution (Clayton, Grossman, and Mayeda 1973). The other anomalies, while perhaps more readily interpretable in terms of processes acting on initially homogeneous material, might also be explained in this way. More generally, many investigators have recently suggested models incorporating pervasive isotopic and chemical inhomogeneities in the early solar system on both microscopic and astronomical scales. No consensus has emerged yet regarding these ideas.

D. Clayton (1975) has extended the presolar dust hypothesis by suggesting that even some isotopic variations which clearly originate in radioactive decay are also carried in interstellar grains. In particular, he suggests that while ^{129}I and ^{244}Pu are in fact

responsible for the meteoritic xenon effects attributed to them, and that while their decay did indeed occur in situ, this decay occurred in interstellar dust grains long before the existence of the solar system, and that these grains were incorporated in meteorites without being vaporized. Conventional conceptions of the chronology of both galactic nucleosynthesis and planetary evolution rest heavily on the assumption of the existence of ^{129}I and ^{244}Pu in the early solar system, rather than merely their fossils. If D. Clayton's hypothesis is correct, it obviously has profound implications for these theories.

There are many approaches by which D. Clayton's hypothesis may be evaluated. We will not review these arguments here but will concentrate on one aspect of the problem, the comparison of ^{244}Pu abundances inferred from xenon measurements and from fission track measurements, which we consider to be an unambiguous test of the hypothesis. To this end we present results of chemical, petrographic, noble gas, and fission track studies of the Allende meteorite, which has played so prominent a role in investigations of early solar system evolution.

II. REFRACTORY INCLUSIONS IN THE ALLENDE METEORITE

The Allende meteorite is classified as a Type III (or C3) carbonaceous chondrite. Allende and other C3 chondrites incorporate white to light-pink inclusions that are usually irregular in shape, occasionally spherical (Clarke *et al.* 1970; Marvin, Wood, and Dickey 1970). Two types of inclusions, coarse-grained (up to millimeter size grains) and fine-grained (micrometer size grains), have been observed. Both are characteristically rich in refractory oxides such as CaO , Al_2O_3 , and TiO_2 . Marvin, Wood, and Dickey (1970) noted that the chemistry and mineralogy of the inclusions was similar to that which Lord (1965) calculated to condense at the highest temperatures from a cooling gas of solar composition. They concluded that the Allende inclusions might represent some of the first solid matter to form in the solar system.

Since these initial observations, considerable effort has been expended in studying the nature and origin of these inclusions, with most work performed on the more tractable coarse-grained inclusions. In general, these studies indicate that the latter inclusions condensed from the solar nebula at high temperatures ($\sim 1400\text{ K}$ at 10^{-4} atm), are enriched in most refractory elements by a factor of ~ 20 over chondritic abundances, and were probably at least partially molten at one time (Grossman 1972, 1973, 1975; Tanaka and Masuda 1973; Wanke *et al.* 1974; Mason and Martin 1974; Blander and Fuchs 1975). The major minerals are melilite (a solid solution between $\text{Ca}_2\text{MgSi}_2\text{O}_7$ and $\text{Ca}_2\text{Al}_2\text{SiO}_7$), clinopyroxene (a complex silicate containing Ca and varying amounts of Mg, Al, and Ti), spinel (MgAl_2O_4), and anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$).

Isotopic studies of these inclusions have aroused substantial interest. The ^{16}O enrichment observed by

Clayton, Grossman, and Mayeda (1973) was first found in Allende refractory inclusions, as were Mg anomalies (Lee and Papanastassiou 1974) and possible evidence for ^{26}Al (Gray and Compston 1974; Lee, Papanastassiou, and Wasserburg 1976). Gray, Papanastassiou, and Wasserburg (1973) found very low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in some of these inclusions, indicating a very early separation of Sr (enriched in the inclusions) from Rb (including ^{87}Rb , which decays to ^{87}Sr). Podosek and Lewis (1973) found a $^{244}\text{Pu}/^{238}\text{U}$ ratio in such inclusions approximately 6 times higher than would have been expected for a chondrite (Podosek 1970a).

The evidence for early formation, the enrichment of refractories (including plutonium and uranium), and a low level of competing interferences combine to make these inclusions especially suitable for study of short-lived nuclides in the early solar system.

III. SAMPLES

The samples of principal interest in this study were obtained from a large coarse-grained inclusion (Fig. 1). Its size (nearly 2 grams) allowed extensive petrographic, track, and noble gas analyses; samples were also reserved for other investigations. This inclusion was separated from U.S. National Museum sample 3666 and provided to us by Ursula B. Marvin of the Smithsonian Astrophysical Observatory; in this work it is designated 3666-I1.

The major minerals in 3666-I1 are spinel (30 vol.%), melilite (36%), and Ti-, Al-rich clinopyroxene (26%); smaller amounts of anorthite (3%), and miscellaneous interstitial material (5%) also occur. This mineralogy most closely fits to type B in Grossman's (1975) classification. A more detailed chemical/petrographic description of this inclusion is given by Shirck (1975); a photomicrograph of a representative section appears as Figure 2.

Noble gas analyses were performed on three fractions of $\sim 0.5\text{ g}$ of matrix-free chips of 3666-I1. All material was crushed to pass 200 mesh ($74\text{ }\mu\text{m}$); material which also passed 500 mesh ($25\text{ }\mu\text{m}$) was collected as a "fines" fraction. The 200–500 mesh fraction was separated into four density fractions by heavy liquids. The fraction with $\rho > 3.33$ was labeled "pyroxene," and the fraction with $2.8 < \rho < 3.2$ was labeled "melilite." The other two fractions had small yields and were not analyzed for noble gases. Material with density $3.2 < \rho < 3.33$ ($\sim 7\%$) had a grain population intermediate to that of the pyroxene and melilite fractions, and material of density $\rho < 2.8$ ($\sim 0.7\%$) was principally plagioclase.

Examination of the density fractions showed an efficient separation of pyroxene and melilite, with spinel and adhering fine-grained material present in both fractions. Unfortunately, uranium distribution measurements (§ V) indicate that in both the whole inclusion and the density separates only about half the uranium resides in the major phases pyroxene and melilite (the spinel is essentially free of uranium), the other half residing in interstitial material. Since the



FIG. 1.—A portion of inclusion 3666-II embedded in the Allende meteorite. Light-colored areas of the inclusion are composed largely of melilite. Dark grains in the inclusion are Ti, Al-rich clinopyroxene.

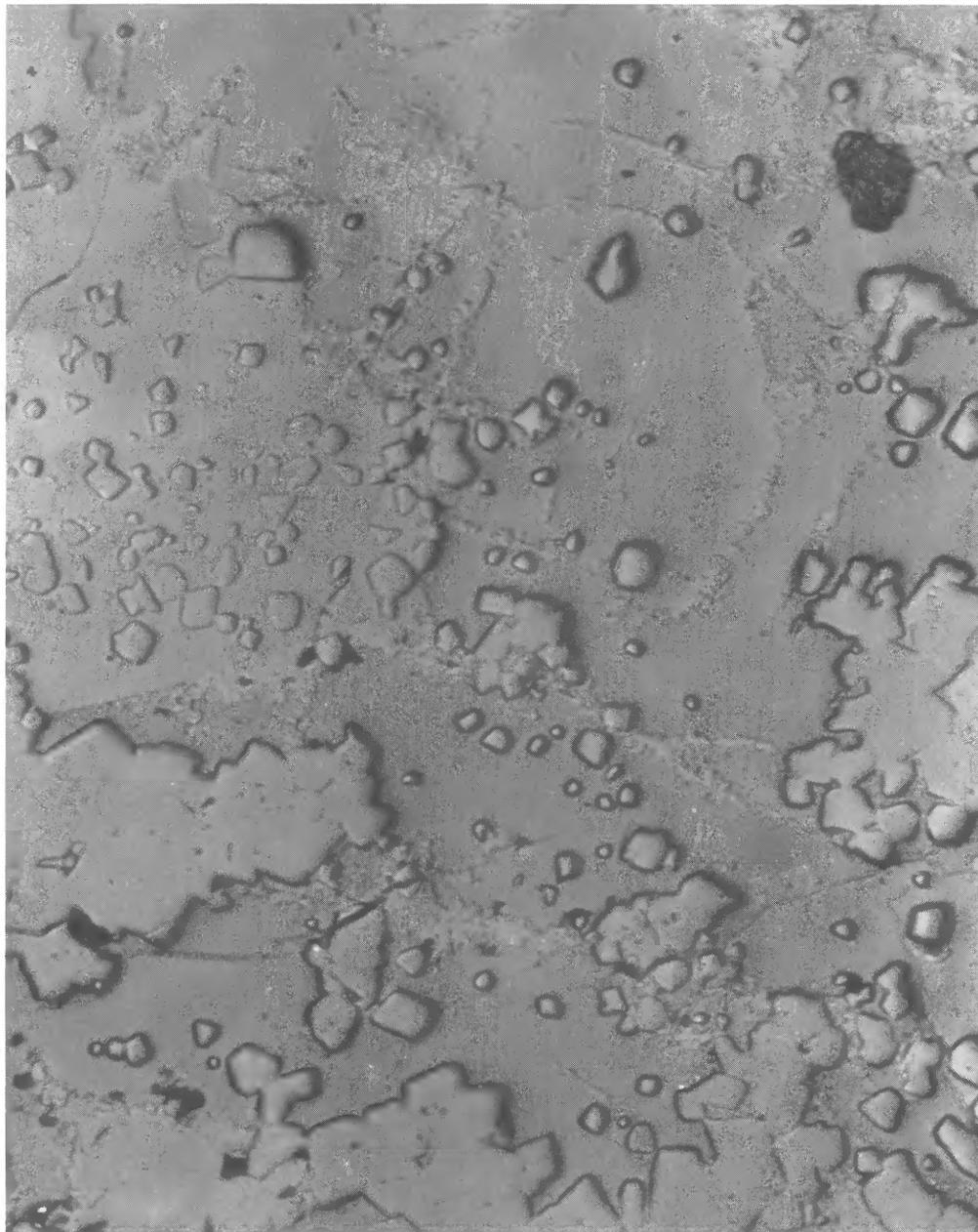


FIG. 2.—A polished surface of 3666-II photographed in reflected light. A lighter colored clinopyroxene grain is visible at the top of the picture. The remainder of the picture is predominantly melilite. The small euhedral grains dispersed throughout the sample are spinel. Stringers of fine-grained material can be seen along the grain boundaries of individual melilite crystals. The field of view is 400 μm . The section was polished differentially to create topographic relief according to mineralogy; the spinel grains, for example, are hardest and thus highest. The dark bands at grain boundaries result from rounding of the edges of raised surfaces.

TABLE 1A
NEON AND KRYPTON* IN ALLENDE SAMPLES†

WEIGHT (g)	SAMPLE	[^{20}Ne]‡			[^{84}Kr]‡				
		cm ³ STP g ⁻¹ × 10 ⁻⁸	$^{21}\text{Ne}/^{20}\text{Ne}$	$^{22}\text{Ne}/^{20}\text{Ne}$	cm ³ STP g ⁻¹ × 10 ⁻¹⁰	$^{80}\text{Kr}/^{84}\text{Kr}$	$^{82}\text{Kr}/^{84}\text{Kr}$	$^{83}\text{Kr}/^{84}\text{Kr}$	$^{86}\text{Kr}/^{84}\text{Kr}$
0.457	Whole rock	5.9§	0.2745 ±0.0004	0.3705 ±0.0006	14.6	0.0495 ±0.0001	0.2030 ±0.0003	0.2006 ±0.0003	0.3130 ±0.0006
0.125	Pink inclusion	2.2#	0.8233 ±0.0016	1.1033 ±0.0016	1.49#	1.47 ±0.10	0.770 ±0.066	0.2087 ±0.0020	0.3116 ±0.0023
COARSE GRAINED INCLUSION 3666-II									
0.120	Pyroxene	2.5#	0.7410 ±0.0016	0.9041 ±0.0023	1.51#	0.0506 ±0.0008	0.2131 ±0.0014	0.2179 ±0.0008	0.3161 ±0.0024
0.081	Melilite	2.5#	0.6077 ±0.0006	0.7713 ±0.0017	0.94#	0.0601 ±0.0021	0.2300 ±0.0017	0.2450 ±0.0029	0.3046 ±0.0022
0.060	Fines	4.6#	0.4151 ±0.0021	0.5391 ±0.0016	1.18#	0.0558 ±0.0011	0.2195 ±0.0030	0.2261 ±0.0024	0.3183 ±0.0030

principal interest in this paper is in plutonium and uranium measurements, the terms "pyroxene" and "melilite" must be understood as labels for the density fractions, and the plutonium xenon and average uranium measurements made on these fractions cannot be considered to characterize accurately these minerals.

Several inclusions besides 3666-II were studied by tracks; the results are discussed by Shirck (1975). Noble gas analyses are reported here for two other samples: whole rock and an interior portion of a fine-grained inclusion, both from U.S. National Museum

sample 4656. The fine-grained inclusion, 4645-I3, also referred to as the "pink" inclusion, has not been quantitatively characterized to the extent that 3666-II has. Nondispersive SEM X-ray analysis indicates the presence of at least four phases: melilite, spinel, nepheline, and sodalite. Because of the fine grain size, no fossil track analyses could be performed.

In addition to the noble gas and track data reported here and in Shirck (1975), other kinds of analyses have been performed on various samples of 3666-II. Rare earth measurements on aliquots of the grain size/

TABLE 1B
XENON IN ALLENDE SAMPLES†

WEIGHT	SAMPLE	cm ³ STP g ⁻¹ × 10 ⁻¹²	[^{132}Xe]‡							
			$^{124}\text{Xe}/^{132}\text{Xe}$	$^{126}\text{Xe}/^{132}\text{Xe}$	$^{128}\text{Xe}/^{132}\text{Xe}$	$^{129}\text{Xe}/^{132}\text{Xe}$	$^{130}\text{Xe}/^{132}\text{Xe}$	$^{131}\text{Xe}/^{132}\text{Xe}$	$^{134}\text{Xe}/^{132}\text{Xe}$	$^{136}\text{Xe}/^{132}\text{Xe}$
0.457	Whole rock	1600	0.00469 ±0.00003	0.00414 ±0.00003	0.0829 ±0.0003	1.797 ±0.004	0.1611 ±0.0002	0.8181 ±0.0011	0.3933 ±0.0003	0.3374 ±0.0002
0.125	Pink inclusion	56§	0.0055 ±0.0003	0.0077 ±0.0002	0.36 ±0.01	414 ±16	0.115 ±0.009	0.662 ±0.013	0.581 ±0.006	0.553 ±0.006
COARSE-GRAINED INCLUSION 3666-II										
0.120	Pyroxene	74§	0.0068 ±0.0003	0.0079 ±0.0004	0.0732 ±0.0010	1.430 ±0.004	0.1383 ±0.0012	0.7475 ±0.0027	0.4818 ±0.0015	0.4467 ±0.0029
0.081	Melilite	67§	0.0076 ±0.0004	0.0107 ±0.0008	0.0812 ±0.0022	2.337 ±0.014	0.1473 ±0.0025	0.7698 ±0.0015	0.4637 ±0.0029	0.4297 ±0.0038
0.060	Fines	95§	0.0070 ±0.0005	0.0081 ±0.0002	0.0796 ±0.0018	1.961 ±0.017	0.1450 ±0.0022	0.7748 ±0.0061	0.4539 ±0.0020	0.4103 ±0.0011

* The ^{78}Kr measurements were compromised by hydrocarbon background in the mass spectrometer and are not reported.

† Data reported are for extraction at 1500° C; subsequent extraction at 1600° C yielded only blank levels of gases. Tabulated errors for ratios are one standard deviation. Ratios have been corrected for instrumental mass discrimination, but tabulated errors do not include contributions of uncertainty in discrimination corrections; these are approximately 0.7%/mass in neon, 0.2%/mass in krypton, and 0.1%/mass in xenon.

‡ Gas concentrations are uncertain by 10% and are reported as measured, without correction for procedural blank. Blank compositions were atmospheric at levels of 4×10^{-10} cm³ STP of ^{20}Ne , 3×10^{-12} cm³ STP of ^{84}Kr , and 4×10^{-13} cm³ STP of ^{132}Xe .

§ Procedural blank between 1% and 10% of measured amount.

|| Procedural blank less than 1% of measured amount.

Procedural blank between 10% and 50% of measured amount.