EVIDENCE FOR THE EXISTENCE OF ^{107}Pd IN THE EARLY SOLAR SYSTEM

William R. Kelly and G. J. Wasserburg

The Lunatic Asylum, Division of Geological and Planetary Sciences, California Institute of Technology, Pasadena, CA 91125

Abstract. The concentration and isotopic composition of Ag and Pd were measured in the Santa Clara iron meteorite. This meteorite has Pd/Ag of about 10^4. The isotopic composition of Pd is identical to terrestrial Pd within 0.1%. The ^{107}Ag/^{109}Ag ratio was 4% greater than the terrestrial value and suggests the *in situ* decay of ^{107}Pd($\tau_{1/2}$ = 6.5 x 10^6y) in this meteorite or its parent body. From the observed ^{107}Ag*/^{110}Pd ratio (* $\equiv$ excess) we infer an initial $(^{107}$Pd/^{110}Pd$)_\odot \geqslant 2$ x 10^{-5}. These data are incompatible with an interval of ~ 10^8 y between the end of nucleosynthesis and the formation of planetary objects but are compatible with a last-minute injection of material. The inferred existence of ^{107}Pd and ^{26}Al indicates that the late injection included freshly synthesized material of both intermediate and low atomic weight on a similar time scale. We estimate the fraction of freshly synthesized material to be between 10^{-5} to 10^{-4} in the region of Z ~ 50. The ^{244}Pu and some of the ^{129}I present in the early solar system may be due to earlier contributions. The highly granular nature of stellar nucleosynthesis is made apparent by these considerations. The ^{107}Pd-^{107}Ag chronometer is of particular importance as it can measure the time interval between the last injection of freshly synthesized material and the formation of differentiated planetary-size objects. Our data suggest that the time scale for the differentiation of some planets into large scale metallic and silicate segregations is as short as ~10^6y.

We report the discovery of an isotopic anomaly in the element silver corresponding to an excess (*) of ^{107}Ag. This anomaly is found in samples of a IVB iron meteorite with a very high ratio of Pd/Ag and is inferred to be due to the decay of ^{107}Pd which has a half life of 6.5±0.3 x 10^6y [*Flynn and Glendenin*, 1969]. After the discovery that ^{26}Al ($\tau_{1/2}$= 0.7 x 10^6 y) was present in the early solar system [*Lee, Papanastassiou and Wasserburg*, 1976, 1977, hereafter LPW; *Papanastassiou, Lee and Wasserburg*, 1977; *Wasserburg, Lee and Papanastassiou*, 1976], it became evident that the time scale for injection of some freshly synthesized nuclear matter into the proto-solar nebula or early solar system was $\lesssim 3$ x 10^6 y before the condensation of refractory phases. This time scale is a factor of ~10^2 shorter than that inferred from ^{129}I ($\tau_{1/2}$= 15.9 x 10^6 y) and raises the basic issue of whether the times are different for last production and injection of low and intermediate Z elements. The question may be phrased in the following manner: Is the observed low abundance of ^{129}I/^{127}I ~ 10^{-4} in the solar system at the time of its condensation due to decay or to dilution? A sensitive and direct test is to determine if there exist isotopic effects from the decay of nuclides in the neighborhood of I but with distinctly shorter half lives. Two such nuclides are ^{135}Cs ($\tau_{1/2}$= 2.3 x 10^6 y) and ^{107}Pd. Variations in the Ba isotopes discovered by *McCulloch and Wasserburg* [1978] include deficiencies and excesses of ^{135}Ba, but no direct correlation with Cs has yet been established. Searches for isotopic anomalies in Ag due to ^{107}Pd decay have been carried out by many workers [*Hess, Marshall and Urey*, 1957; *Murthy*, 1960, 1962; *Chakraburtty, Stevens, Rushing and Anders*, 1964, hereafter CSRA; *Dews and Newbury*, 1966]. The reports of ^{107}Ag excesses [*Murthy*, 1960, 1962] were shown to be in error [CSRA, 1964; *Dews and Newbury*, 1966].

W. R. Kelly and J. W. Larimer proposed that a search be re-initiated in chemical group IVB iron meteorites as condensation calculations indicated these parent bodies condensed at high temperatures relative to other iron meteorite parent bodies and therefore might have very high Pd/Ag ratios [*Kelly and Larimer*, 1975, 1977]. A new search for ^{107}Pd was considered reasonable based on the reduced time scale defined by ^{26}Al and the existence of new microanalytical procedures for Pb which could be adapted to Ag. Following the approach used for the microchemistry of Pb [*Tera and Wasserburg*, 1975], a procedure was developed which permits the determination of isotopic abundances of Ag

to 0.2% on 10^{-9}g of Ag with blanks of 10^{-11}g. This technique was applied to several iron meteorites [*Kelly, Tera and Wasserburg*, 1978, hereafter KTW]. Santa Clara showed a suggestion of an excess of ^{107}Ag of ~ 1%. This meteorite has been subjected to additional analyses including the determination of the Pd concentration and isotopic composition. We note that the precision in measuring Ag isotopic abundance is limited by the ability to control instrumental fractionation.

Experimental Procedures. The procedures used were described in detail [KTW, 1978]. Minor improvements were made in this study and will be noted. All data were obtained on the Lunatic III mass spectrometer. Digital data were obtained with integration times of typically 1 s. The Ag mass region may include additional peaks due to hydrocarbons, Pd$^+$, AsS$^+$, CaPO$_2^+$, and Rh$^+$. The hydrocarbon peaks are typically $\leqslant 1/500$ of ^{107}Ag and are completely resolved with the detector slit width used (0.64 mm). AsS$^+$ produces peaks at masses 107, 108, 109, and 111 with relative intensities of 127:1:5.6:0.02 [see KTW, 1978; in KTW there is an error by the authors; in Tables VI and VII "^{75}As^{32}S$^+$" should read "108"]. CaPO$_2^+$ has a major peak at mass 103 while minor lines occur at masses 105 and 107. Monoisotopic Rh$^+$ also appears at mass 103. Table 1 gives the complete list of Ag and Pd concentration measurements performed in this study. Sample numbers refer to ~5 g slices from a bar of Santa Clara. Isotopic data were obtained at different ion currents and included measurements at the integral mass numbers 103, 105, 107, 108, and 109 and analog scans of the spectrum from 102-110. Background readings were taken ±0.36 a.m.u. from integral mass for all peaks. For integral masses 105 and 108, for which no peaks were observed in the analog output ($< 10^{-17}$ A), the magnetic field settings were calculated based on the carefully centered magnetic field values for masses 103, 107, and 109. In Table 2 data are given for all ratios relative to 109. The first line gives the isotopic ratios at a filament temperature of 1000°C and the second line gives the ranges for the entire run. Limits for 105 and 108 from analog scans were consistent with digital data. The first three samples (1,15,13) were dissolved in HCl only. Centrifugation revealed that these solutions contained a small amount of black precipitate which contained Ag$_2$S. The precipitates were dissolved in aqua regia and treated as separate samples. The combined amounts of Ag were used to determine concentrations in these samples. The Ag concentrations ranged from 0.45 to 2.9 ppb. The first five measurements in Table 2 were suggestive of an isotopic anomaly, but a large unidentified peak at mass 103 was observed. In addition, we were concerned that our dissolution procedure was inadequate and caused variations in the Ag concentrations. From the first three concentration measurements in Table 1 and the corresponding Ag compositions of Table 2 we note there does not exist a strict correlation between the Ag concentrations and the Ag compositions. Large variations in the Pd concentrations do not appear likely. However, there was a clear indication that the Ag concentrations might be variable due to incomplete dissolution of the samples (see Table 1).

Two 0.5 g slices (7.1 and 11.1) were dissolved in aqua regia and totally spiked for both Pd and Ag. There was excellent agreement in the Pd concentrations and the Ag concentrations were in good agreement. We then dissolved slice 11.0 and removed two 300 mg aliquots (11.0.1B; 11.0.1C) which were each spiked for both Pd and Ag. These two replicates gave Pd concentrations in good agreement with each other and the previous analyses. Although the Ag concentrations in these aliquots were in good agreement, they were a factor of 2-3 lower than the previous totally spiked analyses. We were concerned that this might be due to our inability to obtain representative Ag aliquots. Before proceeding further we felt it necessary to address the aliquoting problem and to identify mass 103. In an effort to clarify the first problem, we performed an aliquoting experiment in which we added a known amount of radioactive ^{110}Ag tracer to a 6N HCl solution of 1g of Santa Clara which had been dissolved in aqua regia. Successive aliquots were taken over a 2 week period and were found to contain the correct activity showing that accurate

Table 1. Summary of Ag and Pd Concentration Measurements

	Pd μg/g[c]	Ag ng/g[c]
Santa Clara 1[a] (S 80%, L 20%)[b]	–	0.71±0.03
Santa Clara 15 (L only analyzed)	–	0.45±0.01
Santa Clara 13 (S 14%, L 86%)	–	0.68±0.01 [d]
Santa Clara 7.1A	9.69±0.03	2.4±0.2[e]
Santa Clara 7.1B	–	2.87±0.03
Santa Clara 11.1A	9.72±0.02	2.63±0.07
Santa Clara 11.1B	9.68±0.06	2.60±0.01
Santa Clara 11.0.1B	9.67±0.02	1.06±0.01
Santa Clara 11.0.1C	9.63±0.03	0.89±0.01
Canyon Diablo	–	37.9±0.5
Santa Clara 10A	9.65±0.02	0.97±0.03
Santa Clara 10C	–	0.99±0.01

[a]Slice number . Multiple entries A, B, C represent independent chemical separations of aliquots from the same solution. 11.1 was a small slice cut from 11.0. 7.1 and 11.1 totally spiked for Pd and Ag. 11.0.1B and 11.0.1C were aliquots from the same solution which were each spiked for Pd and Ag. The same is true for 10A; 10C was spiked for Ag only. Errors are 2σ from mass spectrometry measurements. [b]S and L give amounts of Ag in the solid and liquid portion. In samples 1 and 13 these were combined to give total concentrations. [c]Blank corrections applied to Ag data are < 5%. Pd unaffected by blanks. [d]Samples below line dissolved in aqua regia. [e]Large 103 beam observed, data corrected assuming 103 was CaPO$_2^+$. Pyrex plug replaced by quartz fiber after this sample.

aliquoting of solutions ($\sim$100 mg Fe/g) in 6N HCl is possible. Additional mass spectrometer runs indicated that most of the 103 peak in samples 13, 15 and 7.1 was due to CaPO$_2^+$. This problem was greatly diminished by replacing the pyrex plug in the final anion column with one made of high purity quartz. Our revised dissolution procedure caused an increase in the amount of noble metals present in solution which required the introduction of a small cation column to separate these metals from Ag.

Using the above modifications, a 6g slice (10) of Santa Clara was dissolved in aqua regia. Aliquot 10A corresponding to 200 mg of the meteorite was spiked for Pd and Ag (see Table 1) and a 4 g equivalent aliquot (10B) was taken for composition measurements. The initial separation of Fe from Ag was carried out on a 2 ml anion column lg at a time. From the final purified Ag fraction a 3 ng portion (10B.2) was taken for isotopic analysis. The 103/109 changed by a factor of 100 during the run, but there was no concomitant change in the Ag isotopic composition except for a 2% decrease at the end of the run when the Ag

ratio was dominated by fractionation. At 1000°C the 103/105 ratio was $\sim$1000; therefore the 103 beam had a small component of CaPO$_2^+$ and a large component of an unknown peak. We established that Rh does ionize efficiently at 1000°C. The remaining 0.5 ng of Ag (10B.3) was then analyzed. Again there was no variation in the 107/109 ratio with changes in the 103/109,105/109, and 108/109 ratios. Samples 10B.2 and 10B.3 were not run under our standard conditions and therefore we cannot rule out fractionation of $\sim$1%. However, these samples were clearly anomalous and the [107]Ag/[109]Ag ratio was not related to the intensity of other spectral lines. A final modification was a successful effort to remove Rh from the Ag fractions by eluting it from the last column with 1N HCl before eluting the Ag. This procedure was used on a second Ag concentration measurement (10C) and eliminated the dominant contribution at mass 103 (see Table 2). In addition, the Ag concentration was in precise agreement with 10A which was aliquoted and analyzed 1 month earlier. The remaining aliquot (10D) corresponding to 1.2g of Santa Clara 10 was prepared using the modifications in the chemical procedure including the stripping of Rh. The composition of 10D was run under normal operating conditions which included a rigid time schedule for reaching 1000°C and data acquisition on both the Faraday and electron multiplier detectors. These results are given in Tables 2 and 3. From Table 2 we note a substantial decrease in the 103/109 ratio compared to the previous samples due to a decrease in the 103 beam. The previous two runs (B.2,B.3) are fully consistent with the results obtained here using the superior chemical separation and controlled operating conditions.

During every run no correlation was observed between the 107/109 ratio and the intensity of the other spectral lines. It has not proven possible to assign a given spectral line to a specific peak. If one were to assign masses 103 and 105 to CaPO$_2^+$ and mass 108 to AsS$^+$ an assessment of the contributions to the Ag isotopic composition can be made. In the case of 10D the ranges in 103/109, 105/109, and 108/109 would correspond to corrections in the 107/109 ratio of 0.02-0.01%, 0.3-0.07%, and 0.3-2%, respectively. For the 1000°C data the AsS$^+$ correction would be 0.3%. The ranges observed in the 107/109 ratio are not correlated with the other spectral lines, but are compatible with fractionation due to large depletions of the sample during the run. Based on the uniformity of the values obtained on solutions of the same samples under widely varying conditions and the presence of apparent enrichments at mass 107 in several different samples, it is our preferred explanation that these effects are due to an excess of [107]Ag. In particular, sample L13D was a high intensity run with a low 108/109 ratio. It gave a clear indication of a 7% excess in [107]Ag. Insofar as there exist differences in [107]Ag* in this meteorite, it may be possible to determine an isochron. While these data cannot be considered as definitive, they are reasonably interpreted as an

Table 2. Ag Isotopic Measurements on the Santa Clara and Canyon Diablo Meteorites[a]

Sample[b]	[107]Ag Beam (A)	Faraday 107/109	Electron Multiplier 107/109	Electron Multiplier 103/109	Electron Multiplier 105/109	Electron Multiplier 108/109
Santa Clara S1	3x10^{-14}	1.1339±93	1.1456±67	–	–	≤4x10^{-4}
Santa Clara S15	1x10^{-14}	–	1.149±13	–	–	≤8x10^{-4}
Santa Clara L13B	1x10^{-13}	1.149±13 [1.149±13–1.152±9]	–	6x10^{-3} –	3x10^{-4} –	1x10^{-4} –
Santa Clara L13C	1x10^{-13}	1.153±17	1.1653±45 [1.165±4–1.163±7]	6x10^{-2} –	6x10^{-4} –	6x10^{-4} –
Santa Clara L13D	2x10^{-13}	1.1554±17	1.1627±7 –	8x10^{-2} [8x10^{-2}–2x10^{-1}]	–	3x10^{-5} [5x10^{-6}–4x10^{-4}]
Santa Clara 11.0.1A	2x10^{-14}	–	1.145±5 [1.145±5–1.142±17]	0.26 [0.26–0.41]	<5x10^{-4} –	2x10^{-4} [<2x10^{-4}–<7x10^{-5}]
Canyon Diablo	4x10^{-13}	1.0790±26	–	9x10^{-3}	≤1x10^{-4}	≤6x10^{-5}
Santa Clara 10B.2	10^{-13}	(1.128±4)	(1.1395±42) [1.140±4–1.125±40]	0.38 [9x10^{-2}–13]	4x10^{-4} [1x10^{-4}–4x10^{-3}]	8x10^{-5} [8x10^{-5}–3x10^{-4}]
Santa Clara 10B.3	5x10^{-14}	(1.120±14)	(1.1406±70) [1.140±3–1.150±12]	0.92 [4x10^{-1}–22]	1x10^{-3} [9x10^{-4}–1x10^{-2}]	1x10^{-3} [1x10^{-4}–1x10^{-2}]
Santa Clara 10D	10^{-13}	1.1258±41	1.1364±56 [1.141±8–1.128±18]	1x10^{-2} [1x10^{-2}–6x10^{-3}]	4x10^{-4} [2x10^{-3}–1x10^{-4}]	2x10^{-5} [2x10^{-5}–2x10^{-4}]

[a]Samples are listed in chronological order of analysis. All data reported here, including neighboring spectral lines were taken at 1000°C under standard conditions except the numbers in parentheses which were also taken at 1000°C but not under standard conditions. Below the 1000°C data, the total observed ranges are given in brackets. The early runs were not well documented. Errors are 2σ, not 2σ_m. [b]Sample numbers refer to same solution given in Table 1. A, B, C, D denote individual aliquots of these solutions. S, L refer to solid and liquid portions. No corrections were applied to the data.

Table 3. Summary of Ag Isotopic Composition in Normal,
Enriched Standard, and Meteorites[a]

Sample	$(^{107}Ag/^{109}Ag)_F$	$\Delta 107_F$	$(^{107}Ag/^{109}Ag)_M$	$\Delta 107_M$
Santa Clara 10D	1.1258±41 (1)[b]	+4.1±0.4	1.1364±56 (1)	+4.3±0.6
Canyon Diablo	1.0790±26 (1)	−0.2±0.3	−	−
NBS 978 AgNO₃ Normal 2.0%	1.0810±15 (9)	0.0±0.2	1.0895±30 (3)	0.0±0.3
Enriched in [109]Ag	1.0588±58 (4)	−2.0±0.6	1.0677±66 (1)	−2.0±0.7

[a]Errors are 2σ (not 2σ mean). Data taken at 1000°C under standard conditions. Each run ⩾50 ratios. F = Faraday; M = multiplier. $\Delta 107 = [(^{107}Ag/^{109}Ag)_{sample}/(^{107}Ag/^{109}Ag)_{normal} - 1] \times 10^2$. [b]Number of independent runs.

excess in [107]Ag. Table 3 summarizes our Ag measurements taken under standard operating conditions. Santa Clara 10D shows a 4% excess in [107]Ag compared to the metallic phase of Canyon Diablo and to terrestrial Ag. The 4% effect is apparent on both the electron multiplier and Faraday detectors. The discrimination of the multiplier relative to the Faraday is $\sim$1% and follows the $1/\sqrt{m}$ rule. Measurements of a standard 2% enriched in [109]Ag are in good agreement with the gravimetric value of 1.059 and show that a 2% shift in the Ag isotopic composition is readily resolvable during a single determination. We therefore conclude the isotopic data in Table 3 are of adequate reliability with due consideration of fractionation effects and interferring spectral lines. This excess in conjunction with the Pd and the reproducible Ag abundances of sample 10 (see Table 1) yield a concentration of 1.2×10^{11} atoms/g of [107]Ag* in this sample and a ratio of $^{107}Ag*/^{110}Pd = 2.0\pm0.2 \times 10^{-5}$.

It was established that intense ($^{104}Pd \sim 10^{-12}A$) and stable Pd⁺ ion beams could be obtained on 200-500 ng Pd by thermal ionization at 1050°C using the silica gel technique. The Pd concentrations were obtained by both total and aliquot spiking using [102]Pd. Pd was chemically separated in a manner similar to Ag with a total blank of $10^{-10}g$. The isotopic composition of Pd was determined on sample 13C of Santa Clara and on terrestrial Pd (Table 4). The Santa Clara data are indistinguishable from normal Pd to within 0.04% for the major isotopes and 0.2% for the rare isotope. The abundances agree with the earlier results by Sites, Consolœio and Baldock [1953], but are more precise.

Discussion

The $^{110}Pd/^{109}Ag$ ratios for Santa Clara are 250 times greater than found for Canyon Diablo. If $(^{107}Pd/^{110}Pd)_\odot$ were similar to the value $(^{129}I/^{127}I)_\odot \sim 10^{-4}$, then Pd/Ag $\sim$500 would only yield a 1% effect in [107]Ag. All iron meteorites analyzed by CSRA [1964] had Pd/Ag ratios less than 10^2 with the exception of Piñon which had a ratio of 10^3. These samples had normal Ag composition within reported errors. Our results (Table 3) show that the excess in $^{107}Ag/^{109}Ag$ is associated with the sample with a very high $^{110}Pd/^{109}$Ag value. The present data indicate an isotopic anomaly in silver. This anomaly is an excess of [107]Ag (or a depletion of [109]Ag) since the ratio $^{107}Ag/^{109}$Ag is far above the normal value with due regard to instrumental effects. The effect appears to be correlated with the Pd/Ag ratio and is most reasonably associated with the in situ decay of extinct [107]Pd. Two other possible explanations are fractionation effects and effects produced by fission. Arguments have been made for the existence of a chalcophilic superheavy element [Anders, Higuchi, Gros, Takahashi and Morgan, 1975]. A. Turkevich [personal communication, 1976] has indicated that a superheavy element might be siderophilic. The observed effect in Ag could thus be due to in situ fission of a siderophilic superheavy element. The existence of such an

Table 4. Pd Isotopic Measurements[a]

Sample	102/104	105/104	106/104	108/104
Normal Pd[b]	0.09132±5	2.01245±29	2.46077±46	2.39279±57
Santa Clara #13C[c]	0.09155±18	2.01217±59	2.46060±52	2.39247±71

[a]All data were taken on the Faraday detector and normalized to 110/104=1.0604 using a power law. [b]Average of 6 determinations. Errors are 2σ of the means of the 6 determinations. [c]Single determination of 100 ratios. Errors are 2σ (mean).

element has been proposed by Runcorn, Libby and Libby [1977] to provide heat sources for planetary dynamos. Assuming a yield of 1-0.1%, we get a concentration for the hypothetical progenitor of 5-50 ppb. It is also possible that the effect could be due to fission of a known transuranic element in the early solar system.

We will now pursue arguments based on the assignment of [107]Ag* to in situ decay of [107]Pd in the early solar system. We assume further that [129]I and [107]Pd are produced in the same events with comparable production rates. If the value of $^{107}Ag*/^{110}Pd$ is taken as a lower limit for the solar value, we obtain $(^{107}Pd/^{110}Pd)_\odot \geqslant ^{107}Ag*/^{110}Pd = 2 \times 10^{-5}$, from Santa Clara. The most important conclusion from this result is obtained in comparing it with the ratio $(^{129}I/^{127}I)_\odot \sim 10^{-4}$ which is found in all classes of chondrites, enstatite achondrites [Reynolds, 1963; Hohenberg, Podosek and Reynolds, 1967] and recently in silicate inclusions of iron meteorites [Niemeyer, 1978]. If we consider the case of sudden synthesis, which yields the longest time (Δ_{max}) between production (with rates P_i) and planet formation, we have for the observed initial ratio $(N_i/N_j)_o$:

$$(P_i/P_j)e^{-\lambda_i \Delta_{max}} = (N_i/N_j)_o \qquad (1)$$

If the production ratios P_{129}/P_{127} and P_{107}/P_{110} were of the order unity, then as $\lambda_7 \sim 2.5 \lambda_9$, $e^{-\lambda_7 \Delta_{max}} \sim e^{-2.5 \lambda_9 \Delta_{max}} = 2 \times 10^{-10}$. It follows that if [129]I and [107]Pd were made during an event of sudden synthesis 2×10^8 y prior to planet formation, then $^{107}Pd/^{110}Pd$ should be $\sim 10^{-10}$ in contrast to the value of 2×10^{-5} reported here. If the low initial abundance of [129]I were controlled only by radioactive decay subsequent to the last production, then the initial solar system abundance of [107]Pd will be vanishingly small. Using the YONI model of continuous uniform nucleosynthesis [cf. Wasserburg, Fowler and Hoyle, 1960; Wasserburg, Schramm and Huneke, 1969], in which the free decay time is reduced, we have:

$$(P_i/\lambda_i P_j T)e^{-\lambda_i \Delta_{YONI}} = (N_i/N_j)_o \qquad (2)$$

Again using production ratios of the order of unity and a production time (T) of $\sim 5 \times 10^9$ y, it follows that $(^{107}Pd/^{110}Pd)_{YONI} = 1.5 \times 10^{-7}$ which is a factor of 10^2 less than observed. If [129]I and [107]Pd were produced in the same event, the comparable values of $(^{129}I/^{127}I)_o$ and $(^{107}Pd/^{110}Pd)_o$ require that the free decay interval for the last injection must be much smaller than 10^8 y and lead to a model of late injection of freshly synthesized material of intermediate Z [cf. Schramm and Wasserburg, 1970].

For a model of uniform production or a spike (event 1) followed by free decay over a time Δ_1, to which is added a spike (event 2) followed by free decay Δ_2, we have

$$[P_i/\lambda_i \, e^{-\lambda_i(\Delta_1 + \Delta_2)} + P_i \, e^{-\lambda_i \Delta_2}]/[P_j T + P_j] = (N_i/N_j)_o \qquad (3)$$

Here $P_i/\lambda_i, P_j T$ are the number of nuclei present just after long time (T) production, and P_i, P_j are the number of nuclei injected at event 2. From the [26]Al observations and the new results on $^{107}Ag*$, it is now obvious that the short time scale for [26]Al should be extended to other nuclides including [129]I. In this case Δ_2 is $\sim 3 \times 10^6$ y which is negligible for [129]I and small for [107]Pd, and may be set to zero for the present discussion considering the uncertainties in the production rates. For [107]Pd, early production makes a negligible contribution and we have

$$P_i/(P_j T + P_j) \cong (N_i/N_j)_o \qquad (4)$$

This gives the fraction of "i" nuclei made in the last spike. Using the value in Table 3 gives $(N_{107}/N_{110})_o \geqslant 2 \times 10^{-5}$ as $\Delta_2 \geqslant 0$.

If $P_{129}/P_{107} \geqslant (^{127}I/^{110}Pd)_\odot = 7$ this implies that 20% or more of [129]I was added in the last spike. The fraction of [129]I contributed from the last spike is critically dependent on P_{129}/P_{107} for that event. The relative production rates for average "r" process nucelosynthesis have been calculated by Clayton and Fowler [1961] but may not be applicable for a single spike. If, for example, $P_{129}/P_{107} \sim 1$ then only 3% of the [129]I present in the solar system would have come from this last spike.

There remains a question of whether the injected material contained a comparable contribution of U, Th, Pu, and Cm. Arden [1977] has

presented data suggesting U isotopic variations. The abundance of ^{244}Pu ($\tau_{1/2} = 82 \times 10^6$ y) indicates a value of $(^{244}\text{Pu}/^{232}\text{Th})_\odot \approx 5 \times 10^{-3}$ [*Podosek*, 1970]. If all the ^{244}Pu were due to late addition, this would require a yield curve which was a factor of 60 greater in the neighborhood of Z = 94 relative to the solar abundance curve. We consider it more plausible that the ^{244}Pu was dominantly the product of earlier nucleosynthetic events $\sim$ 1-3 x 10^8 y prior to solar system formation in the preceding passages through a spiral arm. The fractional contribution of fresh material (10^{-4} for Z $\sim$50) during the last injection would appear to be rather small. If we consider that all the element injection took place in about 100 passes through the spiral arms, then the production function for nucleosynthesis must be very granular. We may now be resolving in time several distinct periods of element production and injection over the past 2 x 10^8 y. It is possible that the first 10^9 y of production in our galaxy was high enough to account for most of the elements and that the later passages through the spiral arms only produced modest additions. The previous values of Δ_1 obtained for continuous galactic nucleosynthesis from ^{129}I ($\sim$10^8 y) will not change by a large factor if some non-trivial fraction of the observed initial ^{129}I came from "event 1" [*Schramm and Wasserburg*, 1970]. The relative production rates of ^{107}Pd and ^{129}I in the last event are critical parameters in determining the fraction of ^{129}I contributed. The variations in ^{129}I/^{127}I in different meteorites must also be reconsidered as some shifts may be due to inhomogeneous mixing as found for ^{26}Al and not solely due to formation times [*Esat, Lee, Papanastassiou and Wasserburg*, 1978; *A.G.W. Cameron*, personal communication, 1977].

Conclusions

The Ag data are strongly suggestive of an excess in ^{107}Ag due to ^{107}Pd decay in the early solar system, but are not definitive evidence. The Pd-Ag chronometer is unique in that upon melting and segregation of a body to form an iron meteorite large Pd/Ag ratios will be produced in the iron phase. Since in bodies of typical solar system composition the Pd/Ag ratio is about 3, any accumulation of detectable radiogenic Ag will occur only after melting and segregation of the iron phase. The evidence of ^{107}Pd decay in an iron meteorite therefore indicates that the time interval between the last element production and the initial differentiation of a planetary body into iron and silicate phases is commensurate with the half life of ^{107}Pd. Further work will require detailed studies of Pd-Ag systematics both in coexisting phases and in meteorites with different Pd/Ag ratios.

Considering the many isotopic anomalies now observed (O, Ne, Mg, Ca, Kr, Sr, Ag, Xe, Ba, Nd, Sm) and the requirement of a short time scale for both ^{26}Al and ^{107}Pd and consequently for some of the ^{129}I, it is apparent that the "last minute" injection of freshly synthesized nuclear material into the solar nebula covered a large part of the periodic table and was not restricted to low Z elements. We have no *a priori* basis for appealing to separate and special mechanisms to produce ^{107}Pd and ^{129}I independently of one another. The question of whether the last nucleosynthetic event was due to a supernova or was the result of solar system processes in the general sense of *Fowler, Greenstein and Hoyle* [1962] is not resolved.

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