

New data on the abundance of palladium in meteorites

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(Received 19 July 1978; accepted in revised form 10 January 1979)

Abstract—The concentration of Pd in 7 carbonaceous chondrites, 18 ordinary chondrites, 3 achondrites, 29 iron meteorites and other samples has been determined by stable isotope dilution using solid source mass spectrometry. The C1 chondrite Orgueil gives a 'cosmic' abundance for Pd of 1.5 ($\text{Si} = 10^6$ atoms), in good agreement with the currently accepted value.

The concentration of Pd shows little variation among the carbonaceous chondrites, but in ordinary chondrites decreases from the H to L to LL groups. Pd in achondrites is approx 100 times lower than in chondrites. Data for iron meteorites plot around the 'cosmic' Pd/Ni ratio; however the Pd data falls into distinct groups, corresponding to the chemical group classification. These results support the hypothesis that at least two fractionation processes have occurred during the formation of iron meteorites.

1. INTRODUCTION

'COSMIC' abundance tables (e.g. CAMERON, 1973) provide the basic data for theories of nucleosynthesis and cosmo-chemistry. These abundances have been largely based on the Type 1 carbonaceous chondrites (C1), because it is believed that they closely approximate the condensable fraction of primordial solar system material (ANDERS, 1971). Thus the Solar System abundances of the non-volatile elements can be obtained through accurate determinations of their concentration in this class of meteorite.

NICHIPORUK (1971) has reviewed the abundance of Pd in meteoritic material. The Pd data are somewhat variable in quality and many of the replicate analyses, particularly of the carbonaceous chondrites, show considerable spread. NICHIPORUK (1971) concluded that further determinations of Pd in chondrites were required. Only one achondrite has been analysed (HAMAGUCHI *et al.*, 1961), but the abundance of Pd was below the detection limit of the analytical technique used. Very few measurements of Pd in meteoritic material have been reported in the literature since 1971, and CAMERON (1973) therefore used the data on three C1 chondrites reported by NICHIPORUK (1971), to obtain a 'cosmic' abundance for Pd of 1.3 (based on $\text{Si} = 10^6$ atoms). However, this value was obtained by excluding a set of results which were significantly higher than the remaining analytical data. Thus new analyses for Pd in C1 meteorites are needed.

As part of a study of the isotopic composition of Pd in terrestrial and meteoritic material, a solid source mass spectrometric procedure was established which could be used to measure Pd in a variety of samples. Because of the excellent sensitivity, high accuracy and precision of the stable isotope dilution technique, it was considered sufficiently important to analyse a number of carbonaceous chondrites and a

variety of other meteorites, to confirm the 'cosmic' abundance estimate and the distribution of Pd in meteoritic material. Most of the published analyses for Pd have been made by neutron activation, and as far as can be ascertained the isotope dilution technique has never before been used to determine the concentration of Pd in meteorites.

Pd is also an important element in delineating the chemical fractionations which occur in iron meteorites. SCOTT (1972) showed that a number of trace elements formed distinct groups similar to the Ga-Ge groups originally proposed by Brown and co-workers (GOLDBERG *et al.*, 1951; LOVERING *et al.*, 1957) and the chemical groups defined by Wasson and his colleagues (e.g. SCOTT and WASSON, 1975). SUSS (1969) noted that the correlation of Pd with Ni was different from the other noble metals. KELLY and LARIMER (1977) have shown that only Co and Pd condense almost simultaneously with Ni. However, the range of Co concentration in iron meteorites is quite small, whereas Pd varies by a factor of approx seven. Thus Pd is of critical importance in deciphering the cosmochemical history of iron meteorites.

It was therefore decided to determine the abundance of Pd in a representative number of iron meteorites from most of the 12 genetic groups defined by SCOTT and WASSON (1975).

2. EXPERIMENTAL PROCEDURE

The elemental abundance of Pd in each sample was determined using the stable isotope dilution technique. A known quantity of a tracer enriched in mass 102 was added to an accurately weighed sample at the commencement of the extraction procedure. The isotope dilution technique was used to calibrate the tracer against accurate gravimetric standards made up from specpure Pd metal.

Approximately 0.4 g samples of the stone meteorites were analysed. Internal pieces of the meteorite samples were selected in order to minimize terrestrial contami-

Table 1. Concentration of palladium in stony meteorites ($\mu\text{g/g}$)

Sample †	Individual Analyses	Mean	Literature Values
<u>CHONDRITES</u>			
Orgueil C1*	0.61	0.61	0.56 ¹ 0.59 ² 0.53 ⁶
Murchison C2(M)*	0.60	0.63	
Murray C2(M)*	0.66 0.57	0.57	0.79 ⁴
Adelaide #*	0.57 0.59	0.58	
Lance C3(O)*	0.71 0.76	0.73	0.67 ¹ 0.81 ² 1.0 ⁴
Allende C3(V)*	0.60 0.60	0.60	0.70 ⁶
Leoville C3(V)*	0.62 0.67	0.65	
Abee E4*	0.92 0.85	0.88	1.1 ¹ 0.79 ² 0.44 ³
North West Forrest E6	0.62 0.64	0.63	
Yilmia E6	0.67 0.69	0.68	
Brownfield 1937 H3	0.83 0.81	0.82	
Dimmitt H3,4	0.78 0.74	0.76	0.91 ⁴
Wiluna H4,5*	0.96 0.92	0.94	
Allegan H5*	0.82 0.76	0.79	
Cocklebidy H5	0.53 0.56	0.54	
Ijopega H6*	0.83 0.85	0.84	
Gladstone H6	0.78 0.74	0.76	
Saratov L4*	0.61 0.66	0.63	
Dalgety Downs L4,5	0.61 0.61	0.61	
Bruderheim L6*	0.66 0.60	0.63	0.60 ¹ 0.50 ³ 0.68 ⁵
Woolgorong L6*	0.62 0.63	0.62	
Paragould LL5	0.43 0.44	0.44	
North Reid LL5	0.58 0.54	0.56	
Lake Labyrinth LL6	0.50 0.47	0.49	
Mellenbye LL6	0.46 0.52	0.49	
<u>ACHONDRITES</u>			
Millbillillie (eucrite)*	0.009 0.008	0.009	
Nahkla (nahklite)*	0.029 0.018	0.023	
Norton County (aubrite)*	0.025 0.005	0.015	
<u>MISCELLANEOUS</u>			
Tektite (australite)	0.019 0.030	0.024	
Bencubbin (stony-iron)	2.31 0.55	1.43	

* Meteorite fall.

† Classification according to The Revised Cambridge Chondrite Compendium (MOTYLEWSKI, 1978).

DAVY *et al.* (1978) believe Adelaide to exhibit properties of C3(O) and C3(V) types.¹ FOUCHÉ and SMALES (1967).² CROCKETT *et al.* (1967).³ SEN GUPTA (1967).⁴ RIEDER and WÄNKE (1969).⁵ KEAYS *et al.* (1971).⁶ TAKAHASHI *et al.* (1978).

nation. The samples were successively treated with HF, HF-HClO₄ mixture, and 6 M HCl and taken to dryness in a Teflon beaker after each stage of the dissolution. Finally, the sample was redissolved in dilute HCl.

The solution was loaded onto a silica glass column containing Dowex AG1-X8, 100-200 mesh anion exchange resin, precleaned with NH₄OH and equilibrated with HCl. After washing with dilute HCl and distilled water, Pd was eluted with NH₄OH. The eluant was taken to dryness leaving a residue of ammonium salts. Treatment with a HCl-HNO₃ mixture yielded Pd in a form sufficiently pure for mass spectrometric analysis.

Iron meteorite samples approx 1 g in weight, were cut from larger pieces so that inclusion-free pieces could be selected for analysis. The samples were digested with HCl in Vycor flasks. The final solution was adjusted to approx 0.5 M HCl and the Pd extracted as before by ion exchange.

Isotopic analyses were carried out in a 30 cm radius, 90° magnetic sector, solid source mass spectrometer, equipped with an electron multiplier. The samples were loaded on previously outgassed, single, V-shaped, rhenium filaments with silica gel and H₃PO₄. No evidence of Pd contamination from the filament or ion source was observed. An analytical blank was run with each set of samples extracted. Repeated measurements of the blank yielded a value of 1.2 ± 0.2 ng for the complete extraction procedure.

Ion beams of approx 10⁻¹⁴ A. produced from submicrogram-size samples of Pd, lasted several hours. The resulting signals were amplified by an electron multiplier followed by a vibrating reed electrometer with a 10⁹ Ω input resistor. A voltage to frequency converter followed by an electronic counter allowed digital presentation of the data which was fed on-line to a small computer.

To facilitate an internal comparison of accuracy, the mass spectrum was scanned from mass 102 to mass 106. This procedure allowed two concentration determinations of a sample by the isotope dilution technique, using the isotope ratios ¹⁰⁴Pd/¹⁰²Pd and ¹⁰⁶Pd/¹⁰²Pd. Agreement within experimental errors was obtained in all instances.

The mass spectrum in the vicinity of Pd was always examined for the presence of Ru, Cd and hydrocarbons. Ru has isotopes at mass 102 and 104, Cd at mass 106. Both these elements can therefore cause interference to the measurement of the Pd isotopes. Ru was never observed in any of the analyses, but small Cd peaks were occasionally present and corrected for. Hydrocarbon peaks were reduced in size using a liquid nitrogen filled cold finger.

3. RESULTS AND DISCUSSION

3.1 Stone meteorites

The concentration of Pd in 25 chondrites, three achondrites, one tektite and one stony-iron are shown in Table 1. The meteorites were analysed in duplicate to provide an assessment of the precision of the data. The experimental uncertainty associated with each analysis is generally less than 10% at the 95% confidence level. Uncertainties in weight, tracer calibration, blank corrections and mass spectrometric measurements have been taken into account. The only unassessed variable is the effect of inhomogeneities in the samples themselves, and this is undoubtedly a significant source of variation, at least in some samples. The meteorites have been classified according to *The Revised Cambridge Chondrite Compendium* (MOTYLEWSKI, 1978).

The concentration of Pd in the C1 chondrite Orgueil is 0.61 ± 0.10 ppm which is in good agreement with the three literature values listed in Table 1. The large uncertainty in the value of Orgueil is a result of the limited amount of sample (0.08 g) of this meteorite available for analysis. Our value gives a 'cosmic' abundance for Pd of 1.5 (with respect to Si = 10⁶ atoms), using the Si abundance of Orgueil given by WIJK (1969). This compares favourably with the value of 1.3 given by CAMERON (1973). The concentration of Pd in Orgueil can be compared with other 'cosmic' abundances of elements in the same mass region measured in this laboratory using the stable isotope dilution technique (DE LAETER and HOSIE, 1978).

The Pd concentration in carbonaceous chondrites shows little variation. The mean values for the C1, C2, C3(O) and C3(V) meteorites from this work are 0.61, 0.60, 0.73 and 0.63 ppm respectively. The chondrite Adelaide, exhibits properties of C3(O) and C3(V) types (DAVY *et al.*, 1978) and has not been included in either type for the calculation of the previous mean values. The mean value for all C3 meteorites is 0.64 ppm. Previous studies of Pd in carbonaceous chondrites (CROCKET *et al.*, 1967); FOUCHÉ and SMALES, 1967) have shown a small enrichment in the C2 and C3 types relative to C1 chondrites. Our results tend to support this conclusion although further analyses are required to confirm the trend.

The atomic abundances of the C1, C2, C3(O) and C3(V) types are 1.5, 1.2, 1.2 and 1.1 respectively (relative to Si = 10⁶ atoms). With the exception of the C1 value, the Si abundances used are those adopted by MOORE (1971a). Our values indicate that Pd is slightly depleted in C2, C3(O) and C3(V) relative to C1 chondrites, when normalised to Si. Our results differ from those of NICHIPORUK (1971), who reported a constant atomic abundance for Pd in C1, C2 and C3(O) chondrites of 1.3. However in a recent paper, TAKAHASHI *et al.* (1978) reported a slight depletion of Pd in C3(O) and C3(V) chondrites relative to the C1 level, in agreement with our results. This depletion apparently represents loss of metal phase since Fe shows a similar depletion (TAKAHASHI *et al.*, 1978).

The mean abundances of Pd in H, L, LL, E4 and E6 chondrites are 0.78, 0.61, 0.51, 0.88 and 0.66 ppm respectively. The corresponding atomic abundances are 1.2, 0.9, 0.7, 1.4 and 0.9 for the H, L, LL, E4 and E6 chondrites respectively, using the Si abundances of MOORE (1971a). Our results thus support the conclusion of NICHIPORUK (1971) that the atomic abundance of Pd decreases from H to L to LL chondrites. CROCKET *et al.* (1967) reported an atomic abundance from two E4 chondrites of 1.23 which is in good agreement with our E4 value from Abee of 1.4.

The Pd data from this study are linearly correlated with total iron content (MASON, 1971) for the carbonaceous, ordinary and enstatite chondrites, reflecting a constant Pd/Fe ratio during nebular condensation. This correlation is not maintained by the

achondrites. However, a better correlation seems to exist between Pd and Ni (Ni data from MOORE, 1971b) for all stone meteorites, including the achondrites.

The concentration of Pd in achondrites was found to be approximately two orders of magnitude lower than the concentration in chondrites. Pd shows a rough correlation with the Co and Ir values of LAUL *et al.* (1972) and the Ni data of MOORE (1971b) in achondrites. No correlation exists between Pd with Au or Ga (LAUL *et al.*, 1972). However, no definite conclusions can be drawn since the concentration of siderophile elements in achondrites is usually suspect, due to the sporadic distribution of the metallic phase in these meteorites (MOORE, 1971b).

Duplicate analyses of the stony-iron, Bencubbin, did not give reproducible results. Only whole-rock samples of Bencubbin were analysed and the proportion of metal to silicate included in a given sample varied considerably.

The Pd concentration in one australite is also shown in Table 1. As far as can be ascertained this is the first analysis for Pd in tektites. The Pd concentration in the australite is comparable to the concentration in achondrites and terrestrial rocks (DE LAETER and MERMELENGAS, 1978).

3.2 Iron meteorites

The concentration of Pd in 29 iron meteorites is given in Table 2. The meteorites were analysed in duplicate and the associated uncertainties are less than 10% at the 95% confidence level. The Ni content of each meteorite is also given. The values are those compiled by WASSON (1974), with additional data from DE LAETER (1973). The meteorites have been classified using the chemical classification scheme of Wasson and co-workers (SCOTT and WASSON, 1975).

Several determinations of Pd in iron meteorites have been reported previously, with Pd values being available for nine of the irons used in this project. These literature values are shown in Table 2. With the exception of NICHIPORUK and BROWN (1965) who used optical spectroscopy, all previous analyses have been by neutron activation. SMALES *et al.* (1967) determined Pd in 69 irons and obtained a mean value of 4.3 ppm.

The Pd values vary little between the chemical groups, the range for all 29 irons being 1.7–11.3 ppm. The present value of 5.4 ppm for Odessa is higher than the three previously reported values, but this could be the result of sample inhomogeneities. Similarly our value of 2.9 ppm for Sikhote Alin is in disagreement with the estimated value of <1 ppm by NICHIPORUK and BROWN (1965).

SCOTT (1972) has shown that Pd and a number of other trace elements in iron meteorites exhibit strong correlations with Ni, within the chemical groups defined by Wasson and co-workers, thus supporting the genetic significance of the chemical classification. SCOTT (1972) concluded that at least two fractionation events were necessary to produce the observed trace

element distributions. The first process might have occurred during condensation of the solar nebula and produced the fractionation between groups, while the second process, thought to be fractional crystallisation for most groups, produced fractionation within each group. KELLY and LARIMER (1977) attempted to quantify the stages in iron meteorite formation. Their condensation calculations showed that the siderophile elements in the metal phase fall into three broad groups, consisting of elements which have condensation temperatures higher than, similar to, and lower than Ni. The elements Pd and Co were shown to condense almost simultaneously with Ni in the cooling solar nebula, and thus are expected to follow curves that fall close to the cosmic ratio lines.

Figure 1 shows the Pd data, from this work, plotted against Ni content on logarithmic scales. The solid line crossing the diagram represents the cosmic Pd/Ni ratio (CAMERON, 1973). Measurements of the Pd concentration have been made for most of the chemical groups. The data plot around the 'cosmic' ratio line, supporting the hypothesis that Pd exhibits minimal fractionation relative to Ni. However, the separation of the Pd data into distinct groups, corresponding to the chemical group classification, is quite evident. Also, the spread of points within each group is suggestive of a secondary fractionation process. Groups IIAB and IIIAB show clear linear trends and correlate positively with Ni. The situation with respect to the Group IA meteorites is more complex. Three of the four IA meteorites analysed in this study together with Mundrabilla, an anomalous member of Group IA, give a linear correlation with Ni which parallels the 'cosmic' ratio line. The remaining meteorite analysed (Odessa), plots significantly higher, but as already noted, our value is higher than the three literature values listed in Table 2. If the mean of the four values for Pd are averaged, then Odessa would support the concept of a linear correlation confirming the trend noted by SCOTT (1972). This is at variance with KELLY and LARIMER (1977) who have argued that the clustering of the IA data can be explained in terms of a partial melting model. For the remaining groups insufficient data were available to conclude unambiguously that linear trends exist.

SCOTT (1972) showed that the gradient of the observed linear correlation between an element (such as Pd) and Ni is related to the distribution coefficients K_D , by

$$\text{gradient} = \frac{K_D^{\text{Pd}} - 1}{K_D^{\text{Ni}} - 1}$$

where

$$K_D = \frac{C^S}{C^L}$$

and C^L and C^S are the concentrations of the element in the liquid and unmelted solid. SCOTT (1972) assumed a value of 0.9 for K_D^{Ni} and calculated K_D^{Pd}

Table 2. Concentration of palladium in iron meteorites ($\mu\text{g/g}$)

Sample	Classification †	Nickel Content (%)	Individual Analyses	Mean	Literature Values
Canyon Diablo	IA	6.98	4.24 4.12	4.2	3.6^1 5.3^2 4.7^3 2.6^4 3.6^5
Odessa	IA	7.20	5.06 5.69	5.4	4.2^2 3.4^4 3.7^5
Toluca	IA	8.07	4.40 4.68	4.5	3.0^1 4.7^2 5.7^4 4.0^5
Youndegin	IA	6.38	3.97 3.89	3.9	3.9^1
Chihuahua City	IC	6.68	3.77 4.04	3.9	3.9^1
Mt. Dooling	IC	6.26	2.68 2.40	2.5	
Gressk	IIA	5.61	2.09 2.40	2.2	
Bokuslavka	IIA	5.45	1.74 1.74	1.7	
Sikhote Alin	IIB	5.87	2.90 2.85	2.9	$< 1^5$
El Burro	IIB	5.95	2.68 2.84	2.8	
Ballinoo	IIC	9.72	4.55 4.86	4.7	
Kumerina	IIC	9.69	3.70 3.28	3.5	
Carbo	IID	10.02	3.82 4.20	4.0	
Needles	IID	10.3	5.70 6.07	5.9	
Duketon	IIIA	7.52	2.51 2.48	2.5	
Haig	IIIA	7.24	2.69 2.83	2.8	
Henbury	IIIA	7.47	2.55 2.26	2.4	2.1^1 2.0^2 4.7^3 3.9^5
Milly Milly	IIIA	7.62	2.64 3.17	2.9	
Tieraco Creek	IIIB	10.5	7.15 6.77	7.0	
Wonyulgunna	IIIB	8.72	4.08 4.37	4.2	
Carlton	IIIC	13.0	6.86 6.37	6.6	6.7^1 6.5^2
Dayton	IIID	17.02	9.00 9.17	9.1	
Gibeon	IVA	7.68	3.53 3.55	3.5	3.5^1
Mantos Blancos	IVA	8.89	5.60 5.73	5.7	
Tlacotepec	IVB	15.82	6.10 6.11	6.1	
Warburton Range	IVB	17.80	11.39 11.18	11.3	
Mundrabilla	IA-Anom.	7.72	4.62 4.00	4.3	
Mt. Magnet	Anom.	14.56	9.14 9.33	9.2	9.3^1
Redfields	Anom.	6.91	3.77 3.51	3.6	

† Chemical classification according to Wasson and co-workers (SCOTT and WASSON, 1975).

¹ SMALES *et al.* (1967).

² GOLDBERG *et al.* (1951).

³ HAMAGUCHI *et al.* (1961).

⁴ CHAKRABURTTY *et al.* (1964).

⁵ NICHIPORUK and BROWN (1965).

to be 0.65 from the IIIAB group. Our corresponding value for K_D^{Pd} is 0.69. However our K_D^{Pd} value from the IIAB data is 0.28, which is substantially lower. It is not clear whether the discrepancy is due to a real variation in K_D^{Pd} values in different parent bodies or whether it is a result of the accumulation of experimental error in the Pd and Ni concentrations. KELLY

and LARIMER (1977), however, have pointed out that K_D values are expected to vary both with composition and with temperature and have shown that a fractional crystallisation model predicts linear fractionation trends in good agreement with the experimental data. Thus the strong linear correlation of Pd with Ni in Groups IIAB and IIIAB suggests fractional

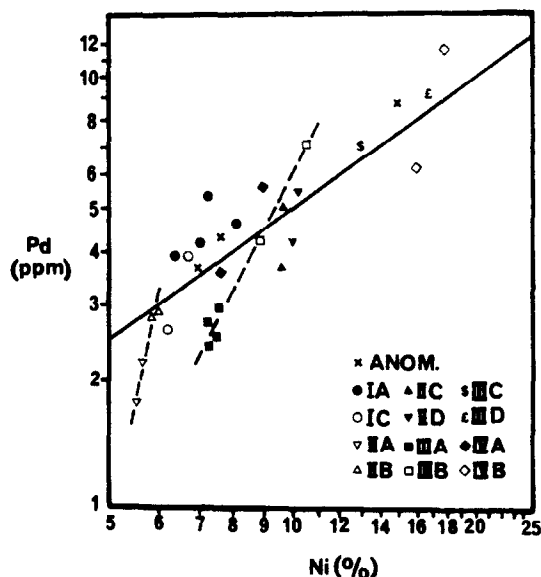


Fig. 1. Pd concentration versus Ni concentration in iron meteorites plotted on logarithmic scales. The solid line represents the 'cosmic' Pd/Ni line (CAMERON, 1973).

crystallisation in a single core. Our data for most of the other groups, although limited, also indicate positive, linear correlations of Pd with Ni.

Our results therefore support the conclusions of SCOTT (1972). In addition our results support the hypothesis that during condensation of the solar nebula, Pd should closely follow the cosmic ratio line. However, it is during condensation that fractionation between groups is likely to occur. A secondary process then caused fractionation within each group and our data support the hypothesis that for most groups fractional crystallisation was the cause of the secondary fractionation.

Acknowledgements—Many of the meteorite samples were generously supplied by the Western Australian Museum Board to whom appreciation is expressed. Professors C. B. MOORE and E. ANDERS kindly provided meteorite samples for analysis. The authors would like to thank Mr. D. J. HOSIE, Mrs. P. R. HARRIS and Miss J. HEPPNER for technical assistance. This project was supported by the Australian Research Grants Committee.

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