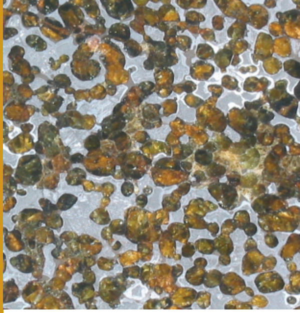
Platinum-Group Elements in Cosmochemistry



Pallasitic meteorite: mixture of olivine and metal resembling the core–mantle region of an asteroid; width ca. 3 cm. PHOTO A. PACK

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In a cooling solar nebula, five of the six platinum-group elements (PGE) condense as refractory-metal alloys at temperatures above the condensation of Fe–Ni metal. Non-refractory Pd condenses in solid solution with Fe–Ni. Such refractory alloys are preserved in some meteorites, although they are often highly altered. The high resistance of PGE to oxidation leads to efficient extraction with metallic Fe–Ni during metal segregation and core formation. Experimentally determined PGE metal–silicate partition coefficients predict lower contents of PGE in planetary silicates than are found, supporting a late addition of PGE components. PGE are particularly useful as tracers of impacting extraplanetary materials in the strongly PGE-depleted crusts of the Earth and other planets.

KEYWORDS: refractory metal, meteoritic component, noble-metal solubility, Ir enrichment

INTRODUCTION

The platinum-group elements (PGE) comprise a group of six rare metals – Os, Ir, Ru, Rh, Pt, Pd – with similar physical and chemical properties (see Brenan 2008 this issue). All six PGE have melting points significantly above the melting point of Fe (1665 K), ranging from 1828 K for Pd to 3306 K for Os (Fig. 2 in Brenan 2008). Metal vapour pressures vary in parallel, with Os having the lowest vapour pressure and the highest melting point of the six PGE. Only two metals, W and Re, have lower vapour pressures than Os.

The term "noble metal" is often used in connection with PGE. There are several definitions of noble metal, such as "any metal that is resistant to corrosion and oxidation" or "a metal whose potential is positive relative to the hydrogen electrode." The second definition includes the six PGE, Cu, Ag, Au and Hg. The resistance of PGE to oxidation is important for understanding their geochemistry. In FIGURE 1, the reduction potentials of the PGE oxides are compared to the reduction potential of FeO. The high values for the PGE reflect the instability of the oxides and the stability of the metals, with Pt oxide being the least stable oxide. All PGE have a strong tendency to partition into metal phases. A quantitative measure is the metal–silicate partition coefficient, $D^{met/sil}_{PGE}$. This is the ratio of the concentration of a PGE in liquid or solid metal to that in silicate melt at given pressure, temperature and oxygen fugacity, and assuming equilibrium between metal and silicate. Elements with partition coefficients above about 10,000 are called *highly siderophile elements* (HSE) and include the PGE, Re and Au. All HSE have very low concentrations in the silicate Earth, reflecting their extraction from the mantle of the Earth with core-forming metal and sulphide (see Lorand et al. 2008 this issue). Of the six PGE, five are classified as *refractory metals*, i.e. their condensation temperatures are above the condensation temperature of Fe–Ni alloys (see FIG. 2). Only Pd is a non-refractory metal, i.e. it has a condensation temperature similar to Fe–Ni (see FIG. 2). Besides the five PGE, the group of refractory metals includes W, Mo and Re.

For a long time, analysis of the PGE was carried out mostly by neutron activation. This method has been particularly effective for Ir, which can be analysed by instrumental neutron activation analysis (INAA) without dissolving the sample and applying radiochemical separation procedures. The other PGE are more than a factor of 10 less sensitive with NAA. Their analysis requires lengthy radiochemical procedures. With the increasingly popular use of inductively coupled plasma–mass spectrometry (ICP–MS), NAA has lost ground and is routinely done in only a few laboratories around the world. Judging from the small number of published analyses, the element Rh may be considered a neglected element compared with the other PGE. Rhodium has no suitable long-lived radioactive isotope for γ counting and it has only one isotope.

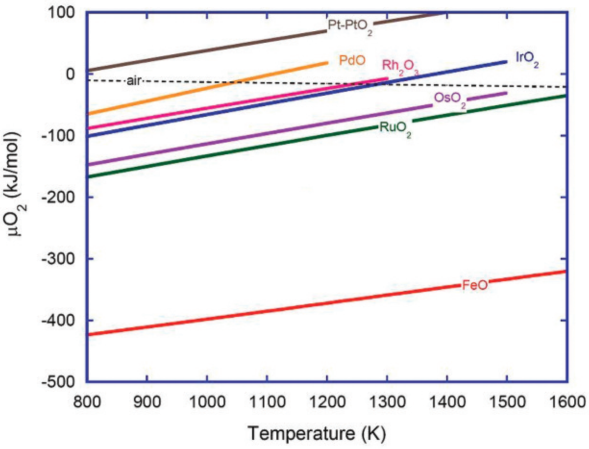

FIGURE 1 Reduction potentials (μO_2) of PGE oxides and FeO as a function of temperature. Data are for equilibrium between pure oxide and pure metal. The reduction potential indicates the thermodynamic stability of oxides compared to metals. The most noble metal is Pt. Its oxide is the least stable of all PGE oxides. FIGURE ADAPTED FROM O'NEILL ET AL. (1995), UPDATED BY J. BRENAN (PERSONAL COMMUNICATION)

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ABUNDANCES IN CI-CHONDRITES AND IN THE SUN

Type 1 carbonaceous chondrites (CI-chondrites) have, within analytical uncertainty, the same chemical composition as the Sun, excluding hydrogen, lithium, carbon, nitrogen, oxygen and highly volatile elements such as the rare gases. Thus they are appropriate reference materials for the average concentrations of PGE in the solar system. The abundances of PGE in CI-chondrite meteorites are around 3 µg/g, corresponding to six PGE atoms for one million Si atoms (TABLE 1). The uncertainties in the meteoritic abundances are around 10% for all PGE (Palme and Jones 2003). For comparison, the abundances of the PGE in the Sun are listed in TABLE 1 (Grevesse et al. 2005). Their errors range from 10 to 30%. The differences between CI-chondrites and the Sun are within the combined uncertainties. As the errors associated with solar abundances are larger than those of CI-chondrites, abundances in the latter should be used for estimating bulk solar system abundances.

The comparatively large differences in the PGE abundances between the Horan et al. (2003) Orgueil meteorite and those in the compilation by Palme and Jones (2003) (TABLE 1) primarily reflect variable absolute contents of refractory PGE in the Orgueil meteorite, which are dependent on the content of inhomogeneously distributed water. Ratios of PGE are less variable.

TABLE 1 PGE ABUNDANCES IN CI-CHONDRITES AND THE SUN

	CI-chondrites			Sun
	concentration (by weight) ng/g		*element/10^6 atoms Si*	
	(a)	(b)	(b)	(c)
Ru	649	683	1.78	2.0
Rh		140	0.358	0.38
Pd	574	556	1.37	1.4
Os	458	506	0.699	0.81
Ir	444	480	0.657	0.69
Pt	867	982	1.32	

Data: (a) Horan et al. (2003); (b) Palme and Jones (2003); (c) Grevesse et al. (2005). Uncertainties: meteorites, around 10%; Sun, from 10 to 30%

BEHAVIOUR OF PGE DURING CONDENSATION IN THE SOLAR NEBULA

The abundances of elements in chondritic meteorites are, to a large extent, determined by their volatility. A quantitative measure of volatility is the 50% condensation temperature. This is the temperature of a cooling gas of solar composition at which 50% of an element is in the solid phase and 50% is in the gas phase, assuming equilibrium between gas and solid. The low vapour pressure of PGE leads to high condensation temperatures. Because of their low abundances and their similar physical and chemical properties, PGE very likely condense as alloys and not as pure elements. In FIGURE 2, the fractions of metals condensing in a common alloy at a given temperature are shown (Palme and Wlotzka 1976; Campbell et al. 2001). The compositions of the condensed alloys can be calculated from the condensation curves and the abundances of the PGE in CI-chondrites.

Five of the six PGE are refractory metals, condensing at higher temperatures than Fe–Ni alloy as refractory metal (RM) nuggets, whereas Pd is a non-refractory metal with a condensation behaviour similar to that of Fe (FIG. 2). This leads to a very different behaviour for Pd compared to the refractory PGE in chondrites and iron meteorites. Unlike the other PGE, palladium is not enriched in refractory components of chondritic meteorites. Also, Pd does not show the large compositional variations that are characteristic of Ir and other refractory metals in many iron meteorites. The similar volatilities of Pd and Fe lead to approximately constant Pd contents in iron and stony meteorites. Radioactive ^{107}Pd, with a half-life of 6.5 million years, decays to ^{107}Ag. Variations in the ^{107}Ag/^{109}Ag ratios of iron meteorites and carbonaceous chondrites are largely due to variable depletions of the much more volatile Ag. The Pd–Ag chronometer thus essentially dates the time of fractionation of volatile elements in the early solar system, with younger ages in iron meteorites resulting from slow cooling and low closure temperatures for Ag isotopes (Carlson et al. 2008 this issue).

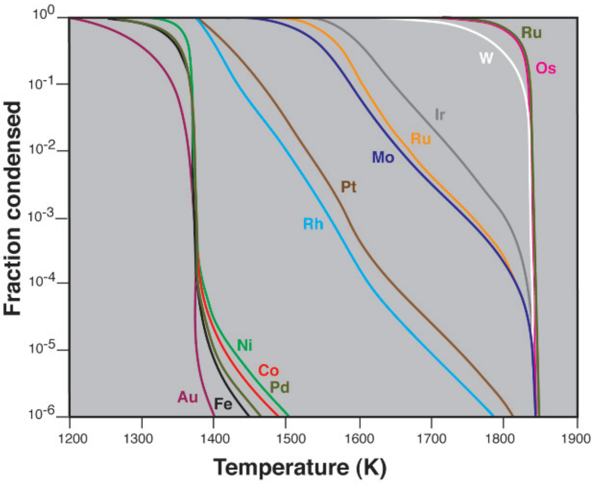

FIGURE 2 Results of condensation calculations. Fractions of PGE condensed at a given temperature are shown (at 10^{-4} atm). Five PGE condense in alloy with W, Mo and Re, but Pd condenses with Fe–Ni alloy. ADAPTED FROM CAMPBELL ET AL. 2001

FIGURE 3 shows the CI-chondrite-normalized pattern of nanometre-sized metal alloys enclosed in refractory meteoritic spinel grains analysed with transmission electron microscopy (TEM) (Eisenhour and Buseck 1992). The agreement with calculated values is striking. The presence of W and Mo in PGE alloys is evidence that such grains formed by condensation in a cooling gas of solar composition. If these RM nuggets were residues of extensive heating and vaporisation, W and Mo would be lost as volatile oxides.

On cooling, initial alloys of refractory metals are modified by exsolution, oxidation and sulfurisation, forming complex opaque assemblages. Such an opaque assemblage with silicates, metal and spinel in a Ca,Al-rich inclusion from the Allende meteorite is shown in FIGURE 4, with secondary phases such as Mo-sulphides (molybdenite), W- and Mo-oxides (scheelite and powellite), Pt,Rh-rich Fe–Ni metal and Os–Ru alloys, the latter two with various amounts of Ir. The bulk grain has a composition perfectly in agreement with condensation calculations, including W and Mo (Bischoff and Palme 1987).

Some of these opaque assemblages look so unusual that El Goresy et al. (1978) initially postulated an origin from outside the solar system and termed them *Fremdlinge,* meaning foreigners in German. Further research has shown, however, that *Fremdlinge* are born in our solar system, primarily because they are isotopically normal (Hutcheon et al. 1987), but the term *Fremdlinge* is still used to describe unusual opaque assemblages in Ca,Al-rich inclusions.

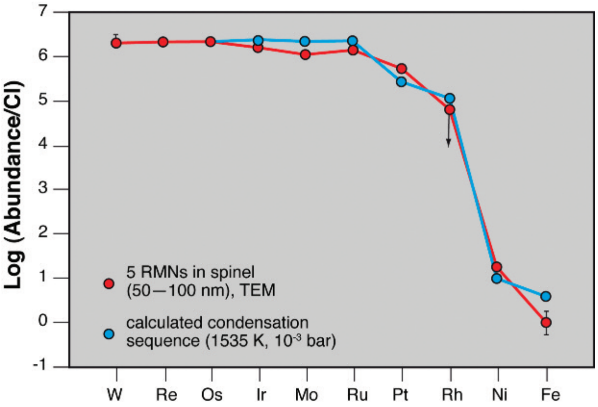

FIGURE 3 CI-chondrite-normalized pattern of nanometre-sized alloys of refractory metals, compared with single-phase condensation calculations. Terrestrial PGE nuggets never contain Mo and W. RMN, refractory metal nugget. FIGURE ADAPTED FROM EISENHOUR AND BUSECK (1992)

PGE IN CHONDRITIC METEORITES

Chondritic meteorites contain variable fractions of a refractory (high-temperature) component with high concentrations of refractory lithophile elements, such as Al, Ca and REE, and also refractory metals. The abundances of all refractory elements are correlated. A good example is a correlation of Al with Ir in various types of undifferentiated meteorites (O'Neill and Palme 1998). The Al–Ir correlation is used when estimating the bulk-planet inventory of refractory PGE (i.e. PGE without Pd). The Ir content of bulk Earth, for example, is derived from the CI-chondrite Al/Ir ratio of $1.77*10^4$, which corresponds to an Ir content for the bulk Earth of 0.91 ppm (O'Neill and Palme 1998). Thus only about 0.2% of the total Ir inventory of the Earth is in the mantle; the rest is in the core (see Lorand et al. 2008).

Recent analyses have confirmed that abundance variations of PGE in chondritic meteorites exceed analytical uncertainties (FIG. 5). Variable ratios of the moderately volatile Pd to the refractory PGE are expected. However, the variability in absolute and relative abundances of the five refractory PGE is comparatively large and cannot be understood in terms of volatility. Some chondrites have high Os/Ir and low Ru/Pt ratios contradicting a simple volatility trend (FIG. 5). Even different meteorites from a single group show variable patterns. Variations in absolute PGE contents in a single meteorite (indicated by the same colour in FIG. 5) reflect inhomogeneous distribution of PGE host phases.

PGE IN PLANETARY MANTLES AND BASALTS

Because of their strong siderophile nature, very low contents of PGE are expected in silicates of differentiated planets. Core-forming metal effectively extracted PGE from the silicate mantles. The comparatively high contents of PGE in the Earth's mantle are often considered to reflect the presence of a late veneer with chondritic PGE patterns. Alternatively, unusually low metal–silicate partition coefficients at very high pressures and temperatures have been invoked (see Lorand et al. 2008). The patterns of PGE in basalts derived from the Earth's mantle are fractionated, with very low Os and Ir and increasing abundances from Ru and Rh to Pd and Pt. This sequence resembles the sequence of increasing crust/mantle abundance ratios (see Fig. 2 of Lorand et al. 2008). The mechanism of fractionation is not known in detail, but likely involves the selective uptake of Os, Ir and Ru, relative to Pt and Pd, in residual olivine, spinel and crystalline monosulphide solid solution.

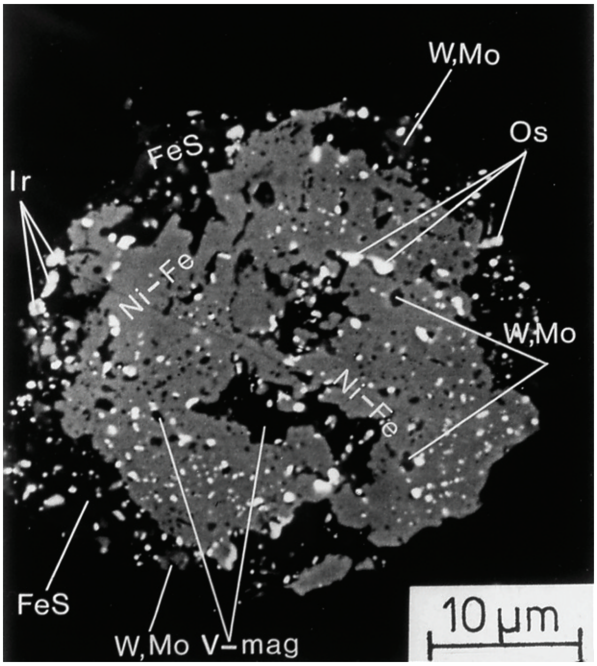

FIGURE 4 Opaque assemblage in an Allende Ca,Al-rich inclusion. Bright spots are Os–Ru alloys with up to 80% Os. On the left side are Ir-rich alloys. Grey phase is Fe–Ni metal with some Pt; the dark phases are powellite, scheelite and V-magnetite. FROM BISCHOFF AND PALME (1987)

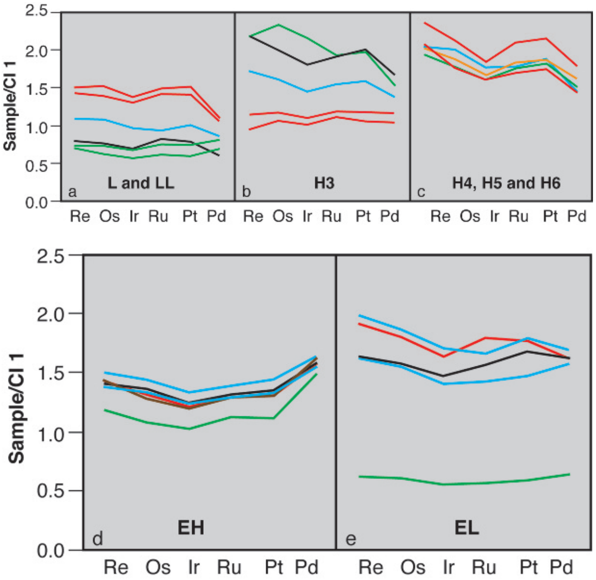

FIGURE 5 The absolute and relative abundances of PGE in chondritic meteorites are variable, certainly beyond analytical uncertainties, which are only a few percent. Each colour designates a different meteorite. Elements are arranged in order of increasing volatility (decreasing condensation temperatures) from left to right, with Pd, the most volatile PGE, at the extreme right. FIGURES ADAPTED FROM HORAN ET AL. (2003)

The PGE pattern in Martian basalts is very similar to that in terrestrial basalts, suggesting similar mantle patterns for both planets (Jones et al. 2003). It is not possible to test this hypothesis as there are no Martian mantle samples available for analysis. If the mantles of Earth and Mars have indeed the same endowment of PGE, Mars may have accreted a late veneer component much like what is assumed for the Earth.

Basalts from the dry (water-free) Moon and the similarly dry asteroid Vesta (the eucrite parent body) have significantly lower PGE contents than basalts from Earth and Mars (Morgan et al. 1978; Righter et al. 2000). The pattern of PGE in the primitive Apollo 12 and 15 basalts is relatively unfractionated compared to that of the more evolved basalts from Apollo 11 and 17, which have more fractionated patterns resembling terrestrial MORB patterns but with one twentieth the absolute PGE contents (Day et al. 2007). The higher than expected PGE contents obtained from partition coefficients (see below), the largely chondritic PGE mantle patterns, and the primitive Os isotope composition suggest that a late chondritic component has been added to Earth, Moon, Mars, and probably also Vesta.

SOLUBILITIES OF PGE IN SILICATE MELTS

The reason for the low concentrations of PGE in silicates from differentiated planets and planetesimals is the strong partitioning of the PGE into metal and sulphides and the subsequent segregation of these phases to form planetary cores. How efficiently can metal extract PGE from silicates? Does the present abundance level in various planetary silicates represent equilibrium between core-forming metal and residual silicates? The answers to these questions require the knowledge of metal–silicate partition coefficients for the PGE. The experimental determination of metal–silicate partition coefficients of PGE relevant to core formation is very difficult. The partition coefficients are very high, and the PGE are only trace elements in Fe–Ni metal equilibrating with silicates during core formation. The task is much easier if equilibrium between pure PGE metals and coexisting silicates is considered. This is essentially the determination of PGE solubilities in silicate melts. Metal–silicate partition coefficients can then be calculated from PGE solubilities. An example is given in Figure 6. Powdered silicates compacted and glued together are suspended in a millimetre-sized Pd-wire loop and heated in a furnace at various oxygen fugacities. The strong decrease in solubility with decreasing oxygen fugacity (Fig. 6) indicates the presence of Pd ions in the silicate melt. The content of Pd in silicate melt increases with increasing oxygen partial pressure. The concave curve can be deconvoluted into a series of straight lines with decreasing slopes, each corresponding to a certain valence of Pd. From these data, a metal–silicate partition coefficient for Pd of $1.5*10^7$ is calculated at 1350°C and an oxygen fugacity appropriate for core–mantle equilibrium (Borisov et al. 1994). The effects of pressure and temperature need to be considered when applying these data to core formation in the Earth (see Lorand et al. 2008).

Other PGE show a similar behaviour. All resulting metal–silicate partition coefficients are extremely high and variable; some are orders of magnitudes greater than those of Pd (Borisov and Palme 1997). Instead, the measured or estimated PGE contents of planetary mantles are much higher than predicted by these partition coefficients, and the patterns predicted would be much more variable if they depended on metal-silicate partitioning.

Either there are other, perhaps kinetically controlled, mechanisms that led to the observed PGE abundance levels in silicates of Earth, Mars, Moon and Vesta, or all of these bodies received a late veneer component with more or less chondritic relative abundances, after silicates had been completely stripped of their PGE during an earlier period of core formation.

One problem with the solubility measurements is that often they do not appear to produce reasonable results under reducing conditions, where the solubilities of Pt and other PGE have been shown to be dominated by the presence of inhomogeneously distributed micronuggets (Borisov and Palme 1997; Ertel et al. 1999). The role of these nuggets in experiments for determining partition coefficients is not clear (e.g. Cottrell and Walker 2006), indicating that the solubilities of PGE in silicate melts are not yet fully understood, despite much experimental effort.

Sulphides are also capable of extracting PGE from silicates. The partition coefficients between liquid sulphide and liquid silicate are expected to be very high (O'Neill et al. 1995). Experimentally determined partition coefficients show a very large range and depend strongly on the composition of sulphides (Fleet et al. 1999; Pruseth and Palme 2004). In addition, the role of sulphur in extracting PGE from planetary mantles is probably minor. After initial separation of S-rich metals, Fe–Ni metal will dominate, as all four solar system bodies we have samples from (Earth, Moon, Mars, Vesta) are depleted in volatile elements, including sulphur. Dreibus and Palme (1996) estimated a maximum S content for the Earth of 0.56%, assuming that Zn and S have similar volatilities. Mars has somewhat more S, but the final stage of PGE extraction will only include Fe–Ni-metal extraction. Moon and Vesta have less S than the Earth.

PGE AS INDICATORS OF METEORITIC CONTAMINATION

The level of PGE in the Earth's mantle is less than one percent of the CI-chondritic abundances (see Lorand et al. 2008). The Ir and Os contents of the crust are a factor of one hundred below those of the mantle. Thus for Ir and Os, there is a difference of four orders of magnitude between their meteoritic and crustal abundances. This makes these

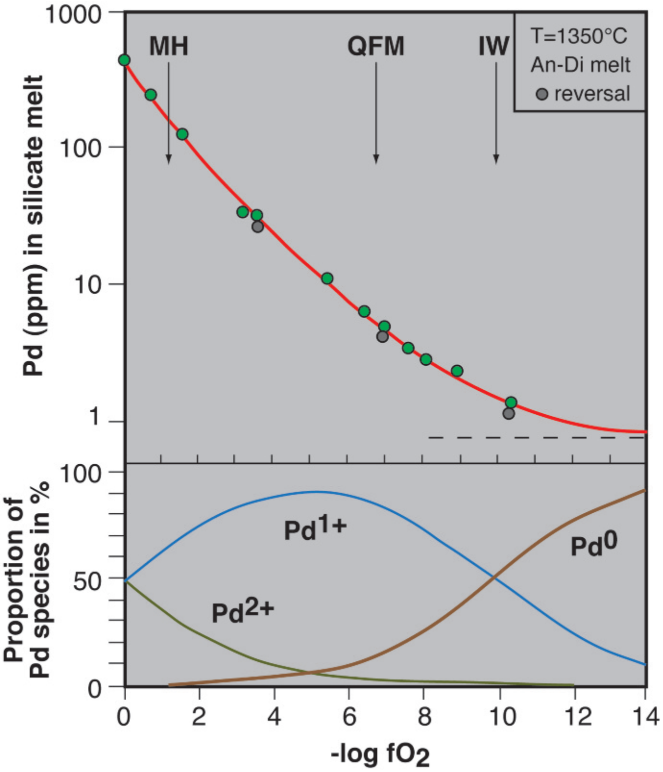

Figure 6 Experimental determination of Pd solubility in silicate melts. A strong decrease in solubility with decreasing oxygen fugacity is observed. In the lower diagram, the contributions of various Pd species to the total number of dissolved Pd ions are indicated. Reversals with high Pd in the initial silicates are indicated. Oxygen fugacity buffers: MH, magnetite–hematite; QFM, quartz–fayalite–magnetite; IW, iron-wüstite. An-Di, anorthite–diopside. Figure modified from Borisov et al. (1994)

elements ideal for studying even minute amounts of extraterrestrial material in impact melts associated with terrestrial craters, as long as the impacting projectile is a chondrite.

A good example is the impact melt sheet at the 20 km diameter Clearwater East crater in Canada (Fig. 7). Based on Ir and Os excesses, more than 5% of a chondritic component was found to be dissolved in the melt rocks at a depth of 320 m below the water level of the lake (Palme et al. 1978). Clearwater East melt rocks have the highest fraction of extraterrestrial component of any terrestrial impact structure. Evidence for PGE enrichment is lacking in many of the 150 impact craters (see review by Koeberl 1998). Low or absent PGE signals indicate that either the projectiles had low PGE contents or the velocity of the impacting object was very high, leading to almost complete vaporisation of the impactor and/or producing such a large amount of melt that the concentrations of dissolved PGE are below the detection limit. Recently, variations in Os isotopes have been used to identify extraterrestrial material. As the $^{187}Os/^{188}Os$ ratio of Earth's crust is much higher than the ratio in primitive meteorites, a small fraction of meteoritic contamination can produce a measurable effect on the $^{187}Os/^{188}Os$ ratio of crustal rocks (Carlson et al. 2008).

A strong meteoritic signature is also present in samples from the K/T boundary layer. Alvarez et al. (1980) first noticed Ir anomalies at the K/T boundary in Gubbio, Italy. Further research showed that (1) the Ir anomaly is present in many marine and terrestrial K/T sections (but not in all) and (2) Ir is accompanied by other PGE metals. The predominance of Ir data in the literature reflects the preference for analysing K/T samples by neutron activation. An example of a terrestrial K/T section is given in Figure 8. The strong decrease in angiosperm pollen coincides exactly with the Ir anomaly. The spore spike reflects a short but severe crisis for land plants, which could arise from lack of sunlight, a prolonged frost period, or acid rain. There is now overwhelming evidence that the Ir and the other PGE at the K/T boundary are of extraterrestrial origin, but it is still unclear if the PGE were transported from the Chixulub crater as meteoritic nuggets, or if the PGE represent condensates from the impact vapour cloud.

CONCLUSIONS

Two properties of the PGE are of particular interest in cosmochemistry. All six PGE (Os, Ir, Ru, Rh, Pt, Pd) have lower vapour pressures than Fe metal, and all six require a higher oxygen fugacity for oxidation than Fe metal. Because of their low vapour pressures, the PGE (except Pd) condense at temperatures above the condensation temperature of Fe–Ni as a common refractory-metal alloy. They condense along with three other refractory metals, W, Mo, Re, and a small fraction of Fe and Ni, but without the more volatile Pd and Au. High-temperature components in meteorites may still contain such alloys, although they may often be sulphurised and/or oxidised. In bulk meteorites, the concentrations of the refractory PGE correlate with those of refractory lithophile elements such as Al and Ca. New data clearly show that ratios of PGE in chondrites are more variable than expected from analytical uncertainties.

During planetary core formation, PGE strongly partition into Fe–Ni metal and are largely removed from the mantle. Experimentally determined solubilities of PGE in silicate allow calculation of metal–silicate partition coefficients. These PGE solubilities are very low and depend on oxygen fugacity, indicating that PGE are dissolved as ions in silicate melts. Calculated partition coefficients between metal and silicate melt are very high, above 10^5 in most cases. The contents of PGE in planetary mantles are higher than

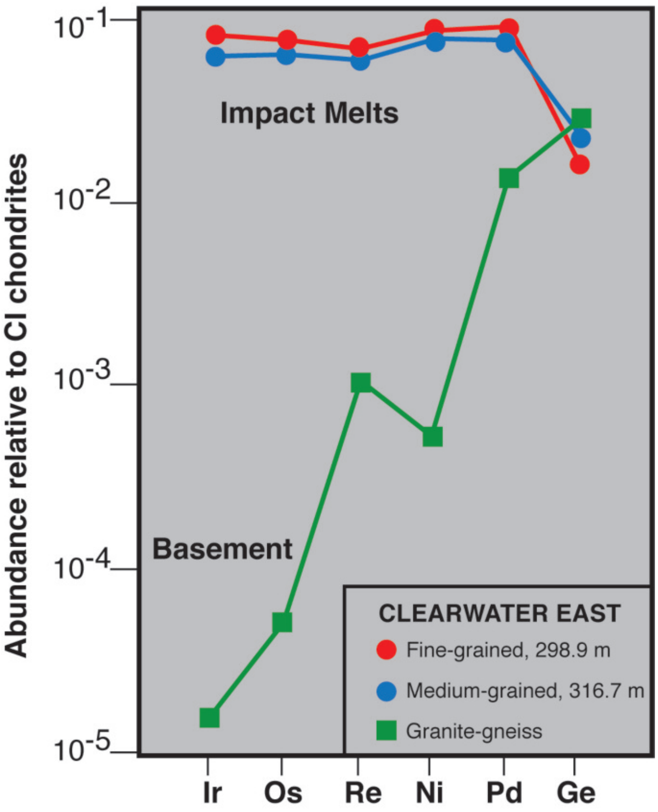

Figure 7 Impact melts from bore holes drilled in winter through a thick ice cover on the 320 m deep Clearwater East Lake, marking a 20 km diameter impact crater in Quebec (Canada). The difference in Ir and Os abundances between meteorites and the Canadian Shield basement is four orders of magnitude. Figure adapted from Palme et al. (1978)

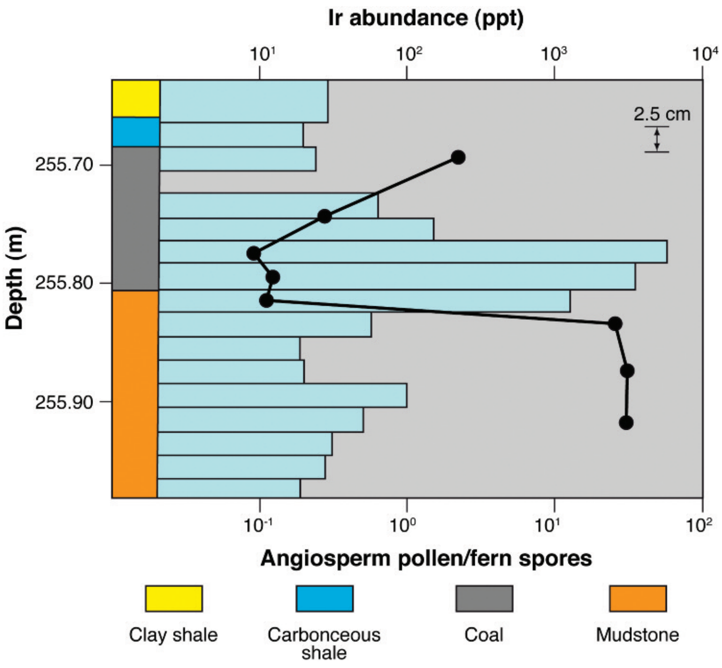

Figure 8 Ir anomaly at a terrestrial K/T section in the Raton basin (Colorado, USA). The strong decrease in angiosperm pollen (solid black circles) coincides exactly with the Ir anomaly (light blue histogram), reflecting severe changes in the environment exactly at the time of Ir delivery. Modified from Orth et al. (1981)

calculated from partition coefficients, suggesting that PGE were added to the mantles of Earth, Mars, Moon and Vesta in the form of a late chondritic component (late veneer) after core formation. Because of the low PGE content of the Earth's crust, PGE are ideal as indicators of extraterrestrial materials in rocks from terrestrial craters formed by impacting cosmic projectiles. The Ir anomaly at the K/T boundary is an excellent example.

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