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Constraints on core composition from shock-wave data

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Seismic data demonstrate that the density of the liquid core is some 8–10 % less than pure iron. Equations of state of Fe–Si, C, FeS₂, FeS, KFeS₂ and FeO, over the pressure interval 133–364 GPa and a range of possible core temperatures (3500–5000 K), can be used to place constraints on the cosmochemically plausible light element constituents of the core (Si, C, S, K and O). The seismically derived density profile allows from 14 to 20 % Si (by mass) in the outer core. The inclusion of Si, or possibly C (up to 11 %), in the core is possible if the Earth accreted inhomogeneously within a region of the solar nebulae in which a C:O (atomic) ratio of about 1 existed, compared with a C:O ratio of 0.6 for the present solar photosphere. In contrast, homogeneous accretion permits Si, but not C, to enter the core by means of reduction of silicates to metallic Fe–Si core material during the late stages of the accumulation of the Earth. The data from the equation of state for the iron sulphides allow up to 9–13 % S in the core. This composition would provide the entire Earth with a S:Si ratio in the range 0.14–0.3, comparable with meteoritic and cosmic abundances. Shock-wave data for KFeS₂ give little evidence for an electronic phase change from 4s to 3d orbitals, which has been suggested to occur in K, and allow the Earth to store a cosmic abundance of K in the metallic core.

1. INTRODUCTION

Constraints on the composition of the Earth's core are inextricably interwoven with theories of the origin, differentiation and thermal evolution of the Earth, as well as coupled to theories of the onset and regeneration of magnetohydrodynamic flows in the liquid outer core. Most of our present knowledge of the Earth is derived from seismic data, which specify both the longitudinal elastic velocity and the density against depth in the outer (liquid) and inner (solid) core of the Earth. The inversion of free-oscillation data, for which certain modes are specifically sensitive to both the elastic constraints and density distribution, as well as the depth of and acoustic impedance contrasts of interfaces inferred from body wave studies, have provided detailed Earth models (see, for example, Dziewonski & Anderson 1981; Hart *et al.* 1977; Gilbert & Dziewonski 1975).

Birch (1952) pointed out upon comparison of seismically derived density–pressure profiles for the Earth and the pressure–density relation for iron (along various thermodynamic paths) that the density of the Earth's outer liquid core is some 8–10 % less than that expected of pure iron over a wide range of hypothetical 'core' temperatures. This inferred deficit in density remains surprisingly little changed if, instead of shock data for pure iron, the Hugoniot data for a meteorite-like composition, *ca.* 6 % Ni – 95 % Fe, or Fe_{0.9}Ni_{0.1} reported by McQueen *et al.* (1970) or Al'tshuler *et al.* (1968), are taken into account (figure 1). The core density deficit

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has generally been attributed to the inclusion of from *ca.* 10 to 30 % (by mass) of various geochemically plausible and cosmologically abundant elements such as Si, S and O, and possibly minor amounts of K, C, N and H.

Jacobs (1975), Brett (1976), Ringwood (1978) and Smith (1977) have reviewed theories of the evolution and composition of the Earth's core, and specific aspects of core composition and formation are discussed by Goettel (1975), Birch (1972), Bukowski & Knopoff (1977), Ito (1976) and Ganguly & Kennedy (1977). Ringwood (1979) has described how different theories of Earth accretion lead to different composition cores. Speculations abound because of the lack of hard data that bear on proposed pressure-induced changes in chemistry of the light elements, specifically S and O, and possibly K.

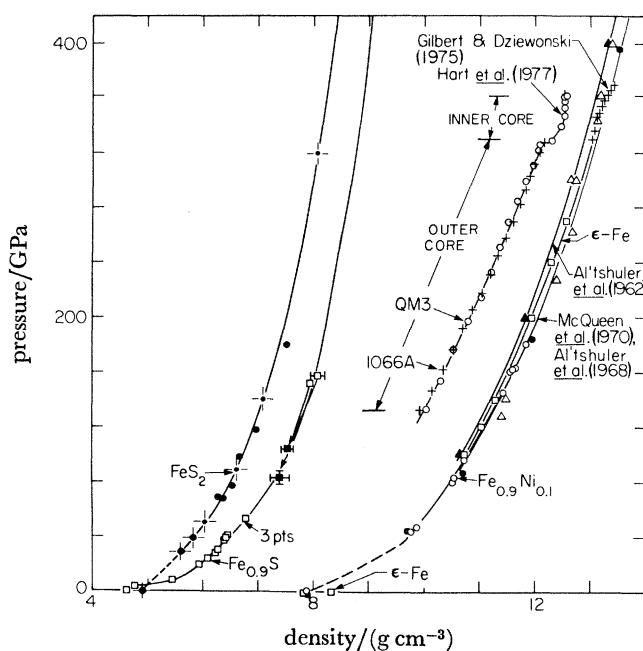


FIGURE 1. Shock pressure plotted against density relations for candidate materials of the Earth's core and seismological data. Shock-wave data for FeS_2 are from Simakov *et al.* (1974) and Ahrens & Creaven, unpublished. Data for $\text{Fe}_{0.9}\text{S}$ are from Ahrens (1979).

The entire range of pressures existing within the Earth's interior, including those present in the outer and inner core, are accessible to the laboratory by using the shock-wave generating technique of projectile impact. This method relies on the launching of metal-bearing projectiles (flyer plates) (composed of tungsten or tantalum, for example) by using, for example, a two-stage light gas gun (Jones *et al.* 1966), which upon impact at speeds of up to 8 km s^{-1} will induce pressures of *ca.* 400 GPa in core materials such as Fe. Moreover, recent shock-wave data for iron place important constraints on the melting point of pure iron under core pressure (Brown & McQueen 1980). The pertinent shock-wave data for the light element core constituents are summarized and discussed in terms of their cosmochemical abundance and the relation of their possible occurrence in the Earth to theories of the formation of the Earth's core.

2. SILICON AND/OR CARBON IN THE CORE

The early, and widely accepted, 'pre-plate tectonics' (for example McDonald & Knopoff 1958; Ringwood 1966) theory for alloying the core involved reducing the silicon in silicates to metallic Si with C and CO in an early reducing Earth atmosphere. As the Earth accreted it subsequently formed a Si-bearing core. This theory suffered from the major problem of strong chemical disequilibrium of the core and mantle. For example, the siderophile elements Ni, Co, Re, Os, Ir, Pt, Cu, Ge, As and Au are too abundant in the mantle for the mantle to have ever been in chemical equilibrium with a Si-rich core. Moreover the mantle contains appreciable Fe^{3+} and the volcanic gases emitted are primarily H_2O and CO_2 . If the mantle had equilibrated with a Fe-Si core, Fe^{3+} would be absent and H_2 and CO would be expected to occur as volcanic

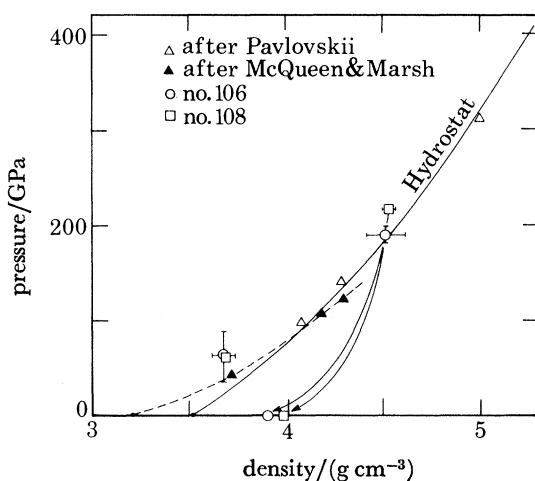


FIGURE 2. Shock-wave data for diamond, and results after Pavlovskii (1971), McQueen & Marsh (1968) and Kondo & Ahrens (1982).

gases. The latter is, however, in accord with the suggestions of Sato (1980), who has pointed out that the occurrence of carbon in the deep interiors of the Earth would lead to a depletion of FeO in the lower mantle. More recently the idea of actually forming refractory carbides and silicides that could be associated with carbon-rich initial condensates such as Fe_3C have been predicted to occur. Larimer (1974) calculated that if the Earth accreted inhomogeneously in a carbon-rich environment such that $\text{C/O} \approx 1$, instead of $\text{C/O} \approx 0.6$ as with present solar photosphere, the initial condensates would be FeNi, minor CaS and TiN and moderate amounts of C, Fe_3C and SiC. This silicide-containing assemblage is found in enstatite chondrites; these meteorites apparently formed in just such a reducing environment. If the Earth's core formed in this way it would be composed of FeNi and contain the high-temperature condensates including silicon and have an appreciable carbon content. Herndon (1979) has also suggested the possibility of such compounds as Ni_2Si and Ni_3Si being in the core.

Maximum carbon contents of the Earth's core may be inferred from isentropes centred at a range of temperatures that may be representative of the core-mantle interface by using Hugoniot data for iron (figure 1) and diamond (figure 2). Densities comparable with that of the outer liquid core of the Earth are compatible with a calculated mixture of iron and 11% carbon by mass. This value for a maximum carbon content is almost independent of

temperature from 3500 to 5000 K and 130 to 328 GPa (the latter represents the pressure range of the outer core). When this (maximum) C abundance for the Earth is cast into the form of a total Earth ratio, C/Si, and compared to solar abundances, the Earth is, at best, depleted in C by at least a factor of about 10 (table 1). Moreover the favourable comparison with carbonaceous chondrites is somewhat misleading because the carbon in these objects is in a highly volatile form of hydrocarbons and carbonates.

TABLE 1. CANDIDATE LIGHT ELEMENT OF THE EARTH'S CORE AND COSMOCHEMICAL ABUNDANCE

light element (X) in the core	Reuss mixtures	percentage by mass in core	(X/Si) atomic fraction, Earth	(X/Si) atomic fraction, chondritic abundance†	(X/Si) atomic fraction, carbonaceous chondrites‡	(X/Si) atomic fraction, solar abundance‡
C	Fe(h.p.p.) + C(h.p.p.)	11	0.44–0.63	0.006	0.7	5.9–14.5
S	Fe _{0.9} S(h.p.p.) + Fe(h.p.p.) also	10–13	0.14–0.29	0.11	0.5	0.24–0.52
O	FeS ₂ + Fe(h.p.p.) Fe _{0.94} O(h.p.p.) + Fe(h.p.p.)	9–13 7–8	3–4	3.5	7.5	13.8–21.4

References: †, Ganapathy & Anders (1974); ‡, Ross & Aller (1976).

Although carbon because of its volatility is precluded from entering the core of the Earth, if the Earth accreted from homogeneous material similar to carbonaceous chondrites, Ringwood (1978) has shown how in the latter stages of such a process large quantities of silicates and iron might be reduced by the carbonaceous material in chondrites and form the hypothetical Fe–Si core material. This is the second mode by which Si, but not C, could enter the core. In both cases, the resulting hypothetical Earth cores are grossly in chemical disequilibrium with the overlying mantle. In the first case a less refractory (more volatile) and oxidized silicate mantle, containing H₂O and CO₂, accreted later, and covered over a reduced and refractory core containing Fe, Si and C. In the second homogeneous accretion case, the oxidized and volatile-rich material initially accreted in the proto-Earth centre is displaced by the liquid, Fe–Si, but not C, core material formed at the surface during the final stages of accretion of chondritic material.

Data for Fe–Si alloys (Balchan & Cowan 1966; Al'tshuler *et al.* 1968) demonstrated that from 14–20% (by mass) of metallic silicon is required to lighten the core. From the point of view of cosmic abundances, a silicon-bearing core yields a total Earth Fe:Si atomic ratio of *ca.* 0.9, in close agreement with the same value observed in carbonaceous chondrites and in the Sun (Fe/Si = 0.71) $\pm_{0.2}^{0.3}$ (Ross & Aller 1976), whereas if the light constituent of the core is not silicon the Earth appears to be decidedly enriched in Fe, with Fe/Si \approx 1.2.

3. SULPHUR IN THE CORE

Sulphur, potassium and sodium are the major volatile elements that appear to be depleted in the Earth's mantle and crust, relative to chondritic and solar abundances. As is demonstrated in table 1, if the outer core is composed of a mixture of iron and the shock-induced high-pressure phases are composed of either FeS₂ or Fe_{0.9}S, S:Si ratios close to meteoritic or solar abundances are inferred.

Recently obtained shock-wave data for FeS₂ (figure 3), when taken with earlier data for

pyrrhotite (figure 1) demonstrate a nearly ideal mixing behaviour for sulphur. Whereas $\text{Fe}_{0.9}\text{S}$ undergoes a phase change probably involving transition from high-spin to low-spin Fe^{3+} , pyrite, which has Fe^{3+} already in the low-spin state, does not appear to undergo a major phase change up to pressures of *ca.* 300 GPa. In addition to sulphur's being present in a plausible cosmic abundance the major attribute of a model Earth's containing sulphur as the major light element in the core is the lowering of the melting point of Fe or FeNi, which at *ca.* 100 GPa is expected to be *ca.* 4000 K, compared with *ca.* 2100 K for the eutectic in the Fe-FeS system (Usselman 1975). Thus considerably lower temperatures are required early in Earth history not only to permit the Earth to retain a moderate inventory of volatiles but also to permit generation of a magnetic field, by core fluid motion, of comparable strength to the present field

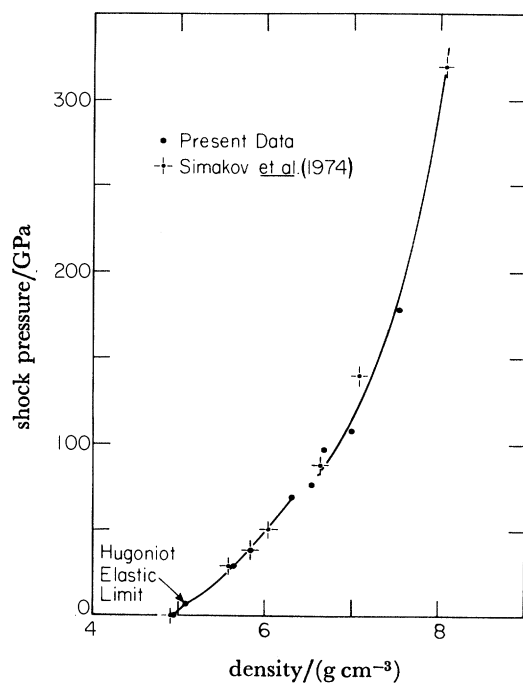


FIGURE 3. Shock-wave data for pyrite (FeS_2). Present data are from Ahrens & Creaven (unpublished).

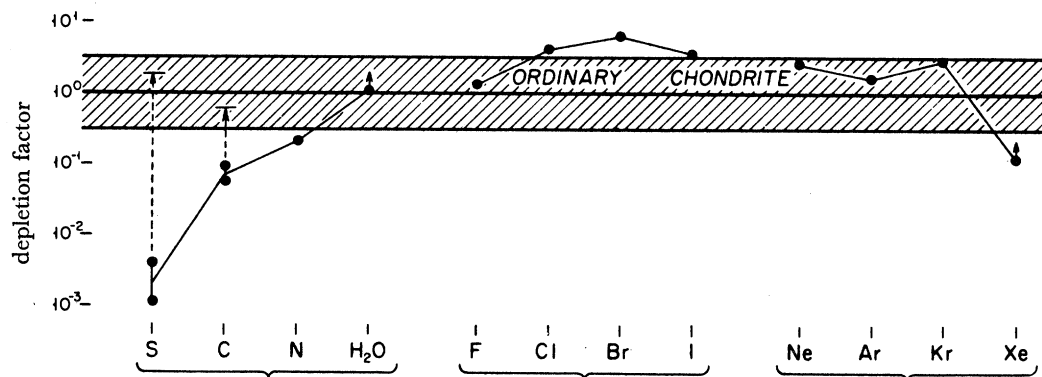


FIGURE 4. Abundances of some volatile elements in the crust plus mantle normalized to their abundance in ordinary chondrites. Broken lines for sulphur and carbon correspond to the amount of these elements missing from the mantle plus crust. Depletion factor = (crust + mantle)/chondritic abundance (after Murthy & Hall 1970).

some 10^9 years after the Earth's formation. The apparent inconsistency of allowing the Earth to accrete nearly its cosmic abundance complement of such volatiles such as F, Cl, Br, I, Ne, Ar, Kr and Xe (figure 4), in addition to S, in contrast to a depletion of K and Na, presents a problem.

4. SHOCK COMPRESSION OF KFeS_2 AND THE QUESTION OF K IN THE CORE

Somerville & Ahrens (1980) have carried out a rather detailed study of the compression of KFeS_2 , a monoclinic compound (zero-pressure density 2.663 g cm^{-3}) to 115 GPa. This work was motivated by the possibility of potassium's becoming a siderophile element and entering the core in small quantities, thereby making the Earth's average composition similar to solar abundances, as suggested by Lewis (1971) and Murthy & Hall (1970). Although it appears that the silicate- K_2S partitioning experiments of Goettel & Lewis (1973), Oversby & Ringwood (1972) and Ganguly & Kennedy (1977) (carried out to 4 GPa) preclude the retention of K in the metallic cores of planets with low internal pressures, e.g. the Moon, the prediction of electronic transitions (from 4s-type bonding to 3d-type bonding, similar to transition metals such as Fe) by Bukowinski & Knopoff (1977) could significantly alter the situation for the larger terrestrial planets, e.g. the Earth. Bukowinski pointed out the significance of ^{40}K with regard to powering the geomagnetic dynamo. Bukowinski & Knopoff's augmented plane wave calculations, even if considered qualitative, imply a very large effective volume decrease, *ca.* 18 % for K. Since K does not, in the sequence calculated by Grossman (1972), condense as feldspar until temperatures of *ca.* 700 °C are achieved, it is expected to 'plate out' on the outside of a homogeneously accreting Earth. Thus to concentrate K in the core from a homogeneously or inhomogeneously accreted Earth, the K-bearing phase must be 'processed', presumably by convection through the high-pressure central portion of the Earth to enter the core.

Ringwood (1978) argues that an electronic phase change in K, about which we know very little, would not necessarily partition K from existing lower-mantle silicates into the core. However, his calculations are very crude and are by no means definitive. Ringwood incidentally points out that Na is also depleted in the Earth's mantle and the above explanation of the apparent mantle K depletion leaves the Na question unanswered. Moreover, other cosmological models suggest that the Earth is in fact *not* depleted in K (Ganapathy & Anders 1974).

As Bukowinski & Knopoff (1977) pointed out, the existing shock data for K (Rice 1965; Bakanova *et al.* 1965) are not definitive with regard to phase changes at *ca.* 30 GPa because of very high shock temperatures. The latter are a consequence of the low impedance of metallic K.

To examine the possible solubility of K in a liquid iron core (figure 5) we calculated the molar volume for K atoms along an isentrope centred at s.t.p. As is evident from the figure, at 100 GPa the atomic volume of K in KFeS_2 is *ca.* $10 \text{ cm}^3 \text{ mol}^{-1}$, whereas $\epsilon\text{-Fe}$ and the high-pressure phases (h.p.ps) of FeS_2 and FeS have volumes between 5 and $6 \text{ cm}^3 \text{ mol}^{-1}$. Hume-Rothery & Raynor's (1962) empirical mutual solubility relations preclude significant solubility of K on these grounds. We concluded, based on the new shock data that we have generated, that K is not likely to be significantly soluble in a liquid iron sulphide core even at core pressures. The possibility of partitioning K into a liquid iron sulphide phase would receive some measure of support if it were shown that KFeS_2 would be stable relative to potassium aluminosilicates at high pressure; on the other hand, a negative outcome does not necessarily dismiss the hypothesis of solubility of K in an iron core. To investigate this question we calculated the effect of

SHOCK-WAVE DATA

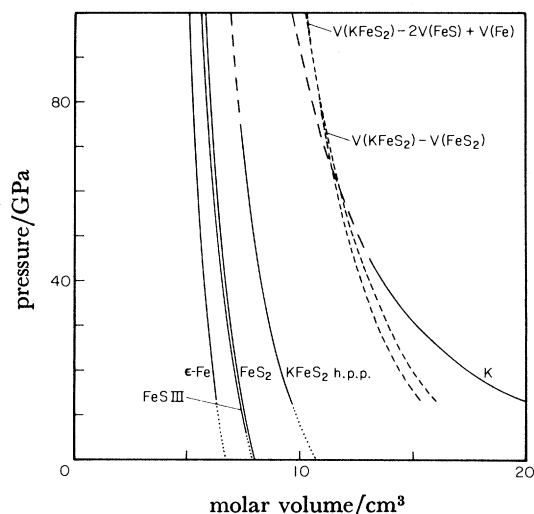
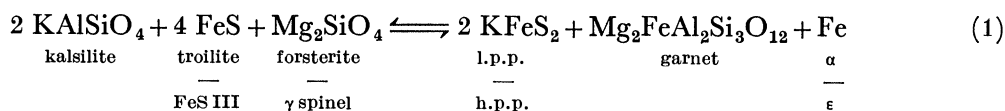
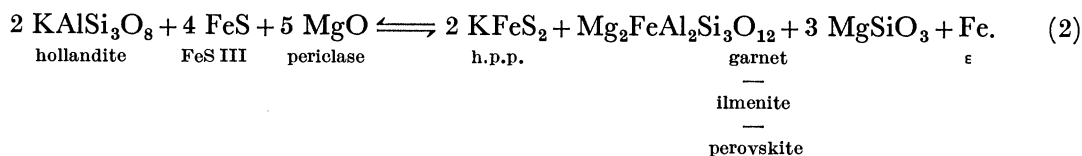


FIGURE 5. Molar volume plotted against pressure for candidate core components (after Somerville & Ahrens 1980).

pressure on the Gibbs energies of hypothetical reactions producing KFeS_2 . The reactions that have been considered are



for pressures up to 20 GPa, and at higher pressures



From thermochemical data we calculated that at pressures above *ca.* 2 GPa, kalsilite (KAlSiO_4) rather than sanidine (KAlSi_3O_8) is the stable potassium aluminosilicate, and that above *ca.* 20 GPa the hollandite phase of KAlSi_3O_8 (Ringwood *et al.* 1967) probably retains the mantle's complement of K. The calculated free energies of hypothetical exchange reactions up to 20 GPa for (1) and, at higher pressures, (2), are strongly positive (by *ca.* 10^5 J/mol), indicating that the high-pressure phase of KFeS_2 is unstable throughout the (assumed low oxygen fugacity) mantle. Moreover, the calculated large positive free energies for the above reactions suggest that if the core differentiated from a nearly homogeneous primitive Earth, it is unlikely that a core-forming liquid iron alloy carried even a trace of K into the Earth's core. Subsequent addition of K to the core at the base of the mantle by exchange reactions, such as (1) and (2), as well as the above-mentioned mutual solubility arguments, appear to be unfavourable toward the possible addition of K in the core during the subsequent history of the Earth.

5. OXYGEN IN THE CORE

The motivation to carry out shock-wave experiments on oxygen, or oxygen-bearing compounds, is not new. Wackerle *et al.* (1968) carried out dynamic experiments on liquid molecular oxygen. They found no evidence for metallization but did not attempt to provide bounds on

the oxygen content of the core. Previous shock-wave data to 1.3 Mbar for Fe_2O_3 ($\rho_0 = 5.27 \text{ g cm}^{-3}$) and Fe_3O_4 ($\rho_0 = 5.20 \text{ g cm}^{-3}$) of McQueen & Marsh (1966) and Syono *et al.* (1977) demonstrate marked phase changes to h.p.ps for which Davies & Gaffney (1973) assigned zero-pressure densities of 6.05 and 6.40 g cm^{-3} respectively. They suggested that the Fe^{2+} and Fe^{3+} in the above minerals had transformed from their normal high spin to a more compact low-spin orbital configuration. Later work by Mao (1974) demonstrated that the phase change in Fe_3O_4 could be observed statically in a high-pressure X-ray apparatus, and displayed the predicted 7% density increase. The spin state in the Fe^{3+} at high pressures was, however, unclear.

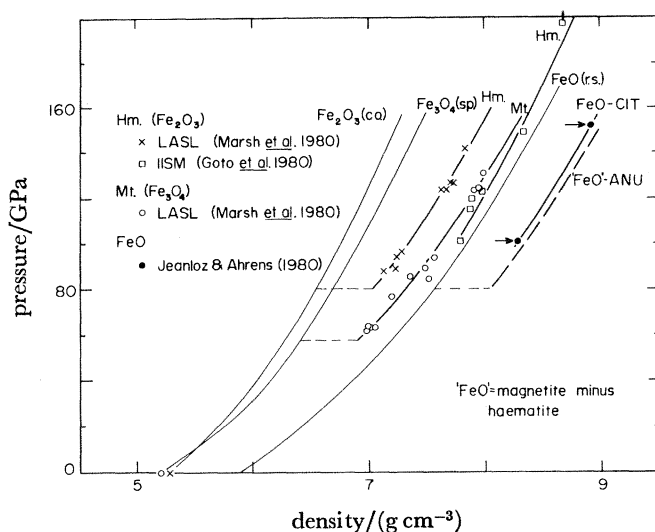
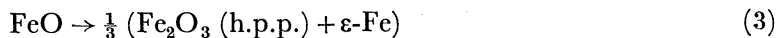
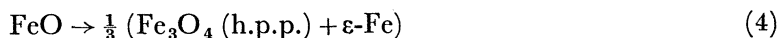


FIGURE 6. Hugoniot data for the iron oxides. The haematite (Hm.) and magnetite (Mt.) curves correspond to linear shock-velocity fits to the data of McQueen & Marsh (1966) (as listed in Marsh 1980) and labelled L.A.S.L.). The recent data of Goto *et al.* (1980) for haematite is also shown as I.I.S.M. The broken curve labelled 'FeO' is obtained by differencing the L.A.S.L. magnetite and haematite Hugoniots. The filled circles represent the $\text{Fe}_{0.94}\text{O}$ data by Jeanloz & Ahrens (1980) corrected to FeO stoichiometry by using the ratio of the corresponding zero pressure densities (the arrows define the extent of this correction). The calculated Hugoniots for the corundum (co.), spinel (sp.) and rock-salt (r.s.) phases are based on third-order Eulerian isentropes. Figure adapted from Jackson & Ringwood (1981).

On the basis of these and the then available data for FeO in the B1 phase, Mao (1974) suggested that reactions such as



and



were (because of the high densities of the h.p.ps) energetically favourable at pressures of *ca.* 20 and *ca.* 30 GPa, respectively. Not only is their experimental evidence that at high pressure Fe^{2+} could oxidize to the Fe^{3+} state, but the above reactions provide a rationale for differentiating iron into the core. Equations (3) and (4) suggest that as the primitive Earth cooled and rapidly convected, its mantle material was, in downgoing convection cells, subjected to pressures of *ca.* 20–30 GPa (depths 600–900 km). Reactions (3) and (4) permitted the FeO component of $(\text{Mg,Fe})\text{SiO}_3$ and $(\text{Mg,Fe})_2\text{SiO}_4$ to disproportionate and removed (metallic) $\varepsilon\text{-Fe}$, which could then sink into a growing core.

Recently Ringwood (1978, 1979) has proposed that the Earth accreted homogeneously from

a mixture of high-temperature devolatilized condensate of the solar nebula (*ca.* 85–90 %) and a carbonaceous chondrite composition (*ca.* 10–15 %) provided that FeO* (metallized) exists and is soluble in liquid iron at high pressures and temperatures. In this case, the primary light alloying element in the core is oxygen, not sulphur, and the core is in chemical *equilibrium* with the mantle. Ringwood discusses high-temperature data that suggest that FeO becomes soluble in Fe (liquid) at high temperature and predicts that the zero-pressure bond length of Fe–O changes from *ca.* 0.216 to 0.201 nm. This implies the existence of a metallic form, FeO*, with a zero-pressure density of 7.34 g cm⁻³, some 24 % greater than stoichiometric wüstite. Electrical resistivity data for magnesio-wüstite, as iron-rich as Fe_{0.56}Mg_{0.44} from 0 to 30 GPa, have not demonstrated this transition (Mao & Bell 1977). Jeanloz & Ahrens (1980) demonstrated a phase change occurring at *ca.* 70 GPa in Fe_{0.94}O and reported shock-wave data to pressures of 175 GPa, although they did not attribute the phase change to metallization. Jackson & Ringwood (1981) suggested that the volume change associated with this phase change is 10–16 % and may be as large as 18–28 %. They propose that the high-pressure phase is either covalently bounded NiAs structure or due to a high-spin to low-spin transition. However, metallization should not be ruled out: the discovery of a very large density increase in quartz to nearly 10 g cm⁻³ at 125 GPa (Palovskiy *et al.* 1978), if true, suggests that we ought to explore the behaviour of silicates such as Fe₂SiO₄.

Although Jeanloz & Ahrens (1980) concluded that the density of FeO at high pressure was too high for reactions (3) and (4) to operate in the Earth, recent shock data for Fe₂O₃ (Goto *et al.* 1980) that indicate that the h.p.p. of Fe₂O₃ is *ca.* 6.31 (compared with the 6.05 g cm⁻³ proposed by Davies & Gaffney (1973)) (figure 6), now appear to allow these reactions.

6. CONCLUSIONS

Shock-wave data for Fe, C, iron sulphides, Fe₂O₃, Fe₃O₄ and FeO can be used to place constraints on the maximum abundances of the light elements Si, C, S and O in the Earth's core. These range from 14–20 % (by mass), for Si, to 7–8 %, for O. Si can enter the core as the result of two very different processes. One mechanism is inhomogeneous accretion in an environment with C/O \approx 1 (compared with C/O \approx 0.6 for the present solar photosphere). In this case a Fe–Ni core will condense first. C will also be present in the form of refractory carbides in this environment. A second type of process would occur during the terminal stages of the homogeneous accretion of the Earth from a chondritic-like composition. Liquid Fe–Si core material could form upon the hot and reducing Earth's surface and sink to the core. The latter process would not yield appreciable C in the core. Another hypothesis is that involving the solubility of oxygen in the core. This suggestion is not inconsistent with the occurrence of major shock-induced transitions in oxides. These transitions, which all occur below 100 GPa in FeO, Fe₃O₄ and Fe₂O₃, may reflect changes in the electronic configuration of Fe. However, recent high-pressure equilibrium data indicate a marked decrease in the solubility of oxygen in the Fe–O–S system with pressure, which makes the solution of oxygen in the core appear difficult. The hypothesis that S (8–12 % by mass) is the light constituent of the Earth's core is consistent with shock-wave data for iron sulphides and implies that the Earth accreted nearly its entire cosmic complement of this volatile element. This is in accord with terrestrial abundance of other volatiles, specifically the halogens and noble gases. Because of the low eutectic melting point in the Fe–FeS system at *ca.* 100 GPa of *ca.* 2100 K compared with 400 K for Fe at this

pressure, the inclusion of S in the core provides a mechanism for producing a molten core early in the Earth's history and retaining the Earth's volatile complement. The Earth's apparent depletion in other major volatiles such as K and Na is not, however, explained.

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