
Chemical Aspects of the Formation of the Core

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Chemical aspects of the formation of the core

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The interpretation of data on lead isotopes in mantle material indicates that some of the lead has been pumped into the core during geological time. Such a continuous lead loss yields an increase of the U/Pb ratio in the mantle.

Quantitative modelling of this effect permits an evaluation of the core's growth curve. In the preferred model, 85% of the core would have been formed in the early period of the Earth's history (50–200 Ma after 4.55 Ga ago) and 15% later on, and its accretion continues.

Some of the consequences of such a process are reviewed.

INTRODUCTION

The Earth's mantle may be considered as a residual material. From the homogeneous matter that accreted to yield the planet Earth, the continental crust segregated toward its surface and the core toward its centre. The composition of the continental crust may be estimated through direct sampling of rocks that outcrop at the surface. The composition of the mantle is estimated from that of basalts and of the few peridotite occurrences at the Earth's surface. The convective circulation of the mantle, indicated by the existence of large lateral displacements of the continents, permits its continuous mixing as a whole, or at least of its uppermost part. Therefore one may consider that basalt samples and peridotite xenoliths are representative of the mantle (upper? whole?). The crust and mantle may thus be directly estimated through rock analyses. On the other hand, the chemical composition of the core may only be inferred indirectly.

Three approaches may be used to infer the core composition:

- (a) a comparison between geophysical observations and high-pressure experiments,
- (b) a study of meteorites and the application of such results to the Earth,
- (c) the chemical budget inferred from estimates for both continental crust and mantle.

Geophysical methods, and more particularly seismic observations and measurements of the Earth's mass and moment of inertia, allow the determination of velocity of seismic waves and density. Laboratory experiments with the use either of shock waves or of the diamond-cell technique allow the study of physical properties of several materials at pressure and temperature conditions close to those in the core. They also yield estimates of the melting temperature of these materials under core conditions. Since the outer core is liquid and the inner core solid, one may thus establish constraints on the actual composition of the core. These two types of observations are discussed elsewhere in this symposium and we shall therefore only recall the essential results.

Nearly everyone now agrees that the core consists essentially of an iron–nickel alloy. However, the density of the core is too low by about 10% compared with that of such an alloy. We must thus admit the presence of 10% light elements in the core. After several controversies discussed by Ringwood (1979) and Stevenson (1981), the debate now focuses on three light elements: O, S and

Si. As suggested by Stevenson, nothing precludes the simultaneous presence of two of these elements, i.e. S and Si or O and Si.

It is worth noting that the presence of a light element decreases the melting point of the Fe–Ni alloy through the formation of a eutectic mixture. The eutectic mixture Fe–FeS has been investigated thoroughly by Usselman (1975) but at pressures still too far below those of the core. He shows, however, that the slope of the solidus is very small and thus, using this eutectic mixture, one could differentiate the core at temperatures low enough to avoid total fusion of the Earth. Moreover, the abundance of ^3He in the mantle would favour the theory of a partly melted Earth (Tolstikhin 1975; Craig & Lupton 1976).

It is worth noting that the FeO–Fe association also corresponds to a eutectic mixture of lower melting point. The effect of silicon in this context and under pressure is still to be investigated. However, it is also quite possible that even a small quantity of P would significantly lower the melting point of the Fe–Ni alloy.

The wide variety of meteorites yields additional information on the internal constitution and formation of planetary objects, and in particular suggest an iron–nickel-rich core. Indeed, non-differentiated objects such as chondrites have mineralogical compositions of the peridotite–native iron type. It is now widely accepted that peridotites are the main component of the Earth's mantle, whence derives the idea that from a primitive Earth, more or less chondritic, an iron core could have segregated, leaving behind a peridotitic residual mantle. This analogy encourages further study of iron meteorites, the texture of which attests to a formation through magmatic segregation of Ni–Fe. Mineralogical observations of these objects show the presence of FeS (troilite), and of siliceous and phosphatic inclusions. The presence of S, Si, P in what some consider as the cores of small planets is thus corroborated.

Trace-element analyses of these objects demonstrate that they have actually been in contact with a silicate part, the extent of which can be inferred from the Ge–Ga distribution.

The discrepancy between the Ga–Ge groups and the classification based on the Ni content suggests that, in the Fe–silicate fractionation, one must take into account the relative abundances of the two components (silicate and Fe–Ni alloy) and also a further parameter, namely oxygen fugacity: the lower this parameter, the greater the tendency of Ni to enter the iron.

A more detailed study of the variations of trace elements in iron meteorites shows that the correlations observed between siderophile elements could be explained by a distillation process (Scott 1971; Drake 1980). If this information were to be applied to the formation of the Earth's core it would have fundamental consequences for models of the process of its chemical segregation.

The budget balance is based on the simple equation

$$\text{core} = \text{bulk Earth} - (\text{mantle} + \text{continental crust}).$$

When dealing with elements in which the continental crust is depleted, we may write:

$$\text{core} \approx \text{bulk Earth} - \text{mantle}.$$

In this method, the main uncertainty is the composition of the bulk Earth. It seems now well established that the concentration of the refractory elements in the Earth is likely to be one to two times that of ordinary chondrites. However, the concentration of volatile elements remains difficult to estimate and we are still far from an agreement between scientists. The method most widely used, i.e. that of choosing a particular type of meteorite as representing the Earth's initial composition, is now subject to controversy.

Let us take as an example the inconsistency that appears when we take C_1 meteorites as representative of the bulk Earth.

Through Nd–Sr correlation, we have a good estimate of the Rb/Sr ratio for the Earth, i.e. about 0.03 (De Paolo & Wasserburg 1976; O’Nions *et al.* 1977; Allègre *et al.* 1979). C_1 type meteorites, often considered as the original material, have ratios 10–20 times higher (Gopalan & Wetherhill 1971). Ordinary chondrites, often used as substitutes, never show such low ratios (Minster & Allègre 1979, 1981). More or less arbitrary mixings of different types of chondrites do not resolve this uncertainty. The differences are attributed to the more volatile character of Rb than that of Sr. In the present state of knowledge, we can only determine limits to the abundance of volatile elements present in the core.

Another approach to this problem is to consider non-volatile elements and to admit for the latter that the analogy with chondrites is valid. In this framework, one may observe that the Earth’s mantle is depleted, not only in siderophile elements such as Ni, Co and Fe, but also in highly siderophile elements such as Au, Pd, Pt, Ru, Rh, Ir, Re and Os. This observation is in full agreement with the idea that the core segregated towards the centre from a homogeneous starting material (Ringwood 1979).

When considering the chalcophile elements usually associated with sulphur, the problem is more difficult to evaluate, since most of them (e.g. sulphur!) are volatile. We are back to the previous problem. We shall thus place the following discussion in the general chemical context essentially based on isotope observations, which allows discussion of the problem of the formation kinetics of the core.

THE LEAD ISOTOPE PARADOX

The lead isotopic ratios $\alpha = (^{206}\text{Pb}/^{204}\text{Pb})$, $\beta = (^{207}\text{Pb}/^{204}\text{Pb})$ and $\gamma = (^{208}\text{Pb}/^{204}\text{Pb})$ vary in Nature. These variations are due to the decay of the isotopes ^{238}U , ^{235}U and ^{232}Th .

With a time-lapse appropriate to the decay constants, the lead isotopic ratios give an estimate of the $^{238}\text{U}/^{204}\text{Pb}$, $^{235}\text{U}/^{204}\text{Pb}$ and $^{232}\text{Th}/^{204}\text{Pb}$ isotopic ratios. When measuring the $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ isotopic ratios in basalts from mid-ocean ridges, it is found that they define a regular evolution line.

In $(\alpha-\beta)$ or $(\alpha-\gamma)$ diagrams, basalts from a large number of oceanic islands fall within this trend towards the most radiogenic domain. For each diagram, one may define a domain of the variations to be expected if the mantle did evolve as a closed system. The equations for this closed system domain are as follows:

$$\alpha = \alpha_0 + \mu(e^{\lambda t_0} - 1);$$

$$\beta = \beta_0 + (\mu/a)(e^{\lambda' t_0} - 1);$$

$$\gamma = \gamma_0 + \kappa\mu(e^{\lambda'' t_0} - 1);$$

where α_0 , β_0 and γ_0 are the initial isotopic ratios at age t_0 (the beginning of the evolution as a closed system), μ and κ are respectively $^{238}\text{U}/^{204}\text{Pb}$ and $^{232}\text{Th}/^{238}\text{U}$, a corresponds to the present value of the $^{238}\text{U}/^{235}\text{U}$ ratio ($a = 137.88$, $\kappa = 3.7$), and λ , λ' and λ'' are the radioactive decay constants of ^{238}U , ^{235}U and ^{232}Th respectively.

One may notice that the trend defined for mid-ocean ridge basalts (m.o.r.b.) lies to the right of the ‘geochron’. This would imply that the evolution did not occur as in a closed system, but that the medium became enriched in U relative to Pb. The geochemical behaviour of Pb and U

is well known: during magmatic differentiation, the melt is more enriched in U than in Pb. Continental crust is also more enriched in U than in Pb during its formation: thus the extraction of continental crust should yield a depletion of U relative to Pb for the mantle, whereas we observe the opposite.

One may call upon the inter-element correlation diagram to characterize this anomaly better. The Nd–Sr isotopic correlation diagram established for oceanic basalts (Richard *et al.* 1976; De Paolo & Wasserburg 1976; O’Nions *et al.* 1977) allows the determination of the $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{87}\text{Rb}/^{86}\text{Sr}$ ratio values for the bulk Earth. These values can be verified through the Sr–Th isotopic correlation diagram (Condomines *et al.* 1981).

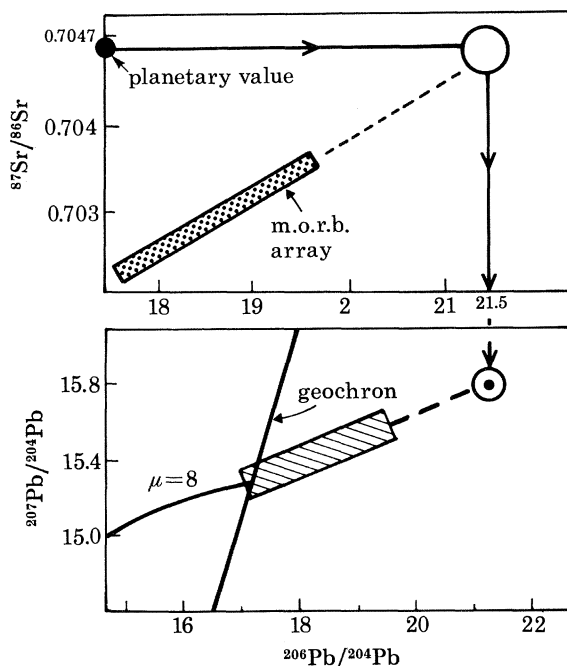


FIGURE 1. Pb–Pb and Pb–Sr correlations for m.o.r.b., which allow determination of the Pb isotopic composition of the bulk Earth from the Sr isotopic composition.

It has also been established that for m.o.r.b. and some ocean island basalts (o.i.b.) (White 1979; Dupré & Allègre 1980; Cohen *et al.* 1980), there is a positive Sr–Pb correlation so that, from the latter, one may determine the Pb isotopic value corresponding to the Sr value for the bulk Earth (figure 1). This value, plotted in either the $(\alpha-\beta)$ or $(\alpha-\gamma)$ diagram falls outside the domain for closed-system behaviour and to the right of the geochron.

The abnormal chemical properties of U and Pb relative to those of Rb–Sr and Sm–Nd, which is referred to as the lead paradox (Allègre 1969; Allègre *et al.* 1980), may be interpreted as the result of Pb being pumped into the core from the mantle through geological time. The sulphur-containing core incorporates lead but not uranium, so that its μ value is zero and it represents an ideal system to increase the μ value of the mantle throughout geological time. In this respect, the o.i.b. originating from the deep mantle will be more affected than the m.o.r.b. derived from a more superficial mantle.

The other alternative, suggested by O’Nions *et al.* (1979), is that the low μ reservoir is the lower continental crust. This can only work if the whole continental crust has a lower μ value than the

mantle. However, the Pb isotopic data on the evolution of the mantle–crust system throughout geological time do indicate that

$$\mu_{\text{continental crust}} > \mu_{\text{mantle}}$$

Another possibility (Hofmann & White 1980) would be to reinject highly U-enriched sediments into the deep mantle. We should then obtain highly radiogenic values for o.i.b. and lower radiogenic values for m.o.r.b. But such a scheme would be in complete disagreement with $^3\text{He}/^4\text{He}$ ratio data, which show that o.i.b. are more enriched in ^3He (and therefore less enriched in radiogenic ^4He) than m.o.r.b.

In conclusion, in the present state of knowledge, the model of Pb pumping by the core seems to be the most likely and we shall now describe this model, which has already been discussed elsewhere (Allègre *et al.* 1980; Dupré & Allègre 1980). Our idea differs from that proposed by Vidal & Dosso (1978), who suggest the same process with simultaneous Rb pumping, and from that proposed by Volmer (1977) in which the core is involved in this process for a very limited time.

QUANTITATIVE MODEL FOR LEAD ISOTOPIC EVOLUTION

A model for the evolution of the lead isotopic ratios cannot be derived independently from the structure of the mantle and from its assumed evolution. Thus we have to consider two extreme models with two purposes in mind:

- (a) a quantitative explanation of the lead isotopic composition of the mantle, and
- (b) the inferring of a growth curve for the mantle.

One may write the general equations for the evolution of the U–Pb system (Allegre *et al.* 1980) as

$$d\alpha/dt = \lambda r;$$

$$dr/dt = (\psi + \psi_c - \lambda) r;$$

where $\psi(t)$ is the fractionation factor between the continents and the mantle and $\psi_c(t)$ is the fractionation factor between the core and the mantle; r is the parent/daughter isotopic ratio.

Similar equations can be written for β and γ with the same ψ and ψ_c but with different λ .

The ψ_c values will be of two types. For the early days, when the primary differentiation of the core occurred, ψ_c is considered to have been large and constant; then, after this period, ψ_c is taken to have decreased according to the following exponential law of decay:

$$\psi_c = \psi_0 e^{-t/\tau}.$$

The integration of these differential equations throughout the Earth's history requires a certain number of assumptions. These assumptions are linked to the mantle model considered. The two extreme schemes used correspond respectively to a whole-mantle convection model and to a two-layer model.

(a) Whole-mantle convection model

We suppose that at an early stage of the Earth, the core segregated suddenly. Then, after this initial stage, the rate of core formation decreased exponentially with time.

At 4 Ga B.P. the continental crust started to form, extracting U and Pb out of the mantle. However, the U/Pb fractionation of such a process is not very large and therefore, since the continental crust represents only a small part of the mantle, this mechanism is not very important for U/Pb fractionation. Within this general model, we have considered two extreme cases.

Case A. The primary differentiation of the core yields a heterogeneous mantle, more depleted for some domains than others. Then, when convection starts, ψ_c is the same for the whole mantle.

Case B. The initial segregation of the core occurs during a convective régime yielding a homogeneous mantle with a constant μ . Afterwards the segregation of the core is more efficient in some domains close to the core–mantle interface than in the surface domain and thus the ψ_c values are variable.

For the sake of simplicity, we assume that the primary differentiation of the core occurred during the first 200 Ma of the Earth's history; though such a process could have occurred during

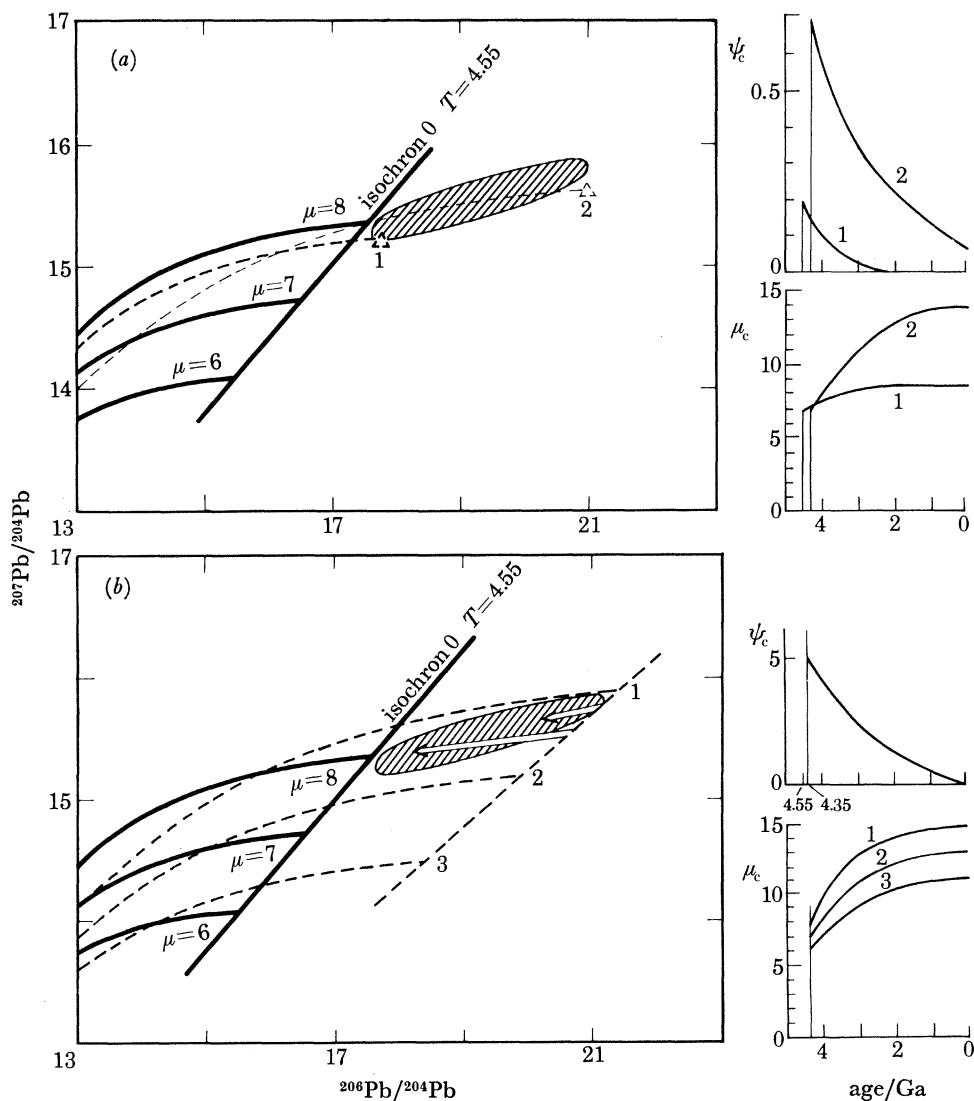


FIGURE 2(a) AND (b). For description see opposite.

the first 50 Ma only, this would yield nearly identical results. Because of the low resolution obtained for the lead isotope data, we have taken a maximum duration for this differentiation.

Numerical calculations have been done for several cases with various possible μ_0 values, μ values after the primary differentiation and different $\psi_{0,c}$ and τ values (time constant for ψ_c decay).

As illustrated in figure 2, several scenarios can explain the lead isotope distribution observed for the mantle and thus for basalts. The effects of core extraction on several parameters are illustrated.

(b) *Two-layer mantle model*

In such a model, the mantle is divided into two parts: the upper mantle, which is the source for m.o.r.b. and which corresponds to a layer 500–1000 km thick; and the lower mantle, which is the source for o.i.b. and for hot spots, and which corresponds to the part close to the core. We suppose

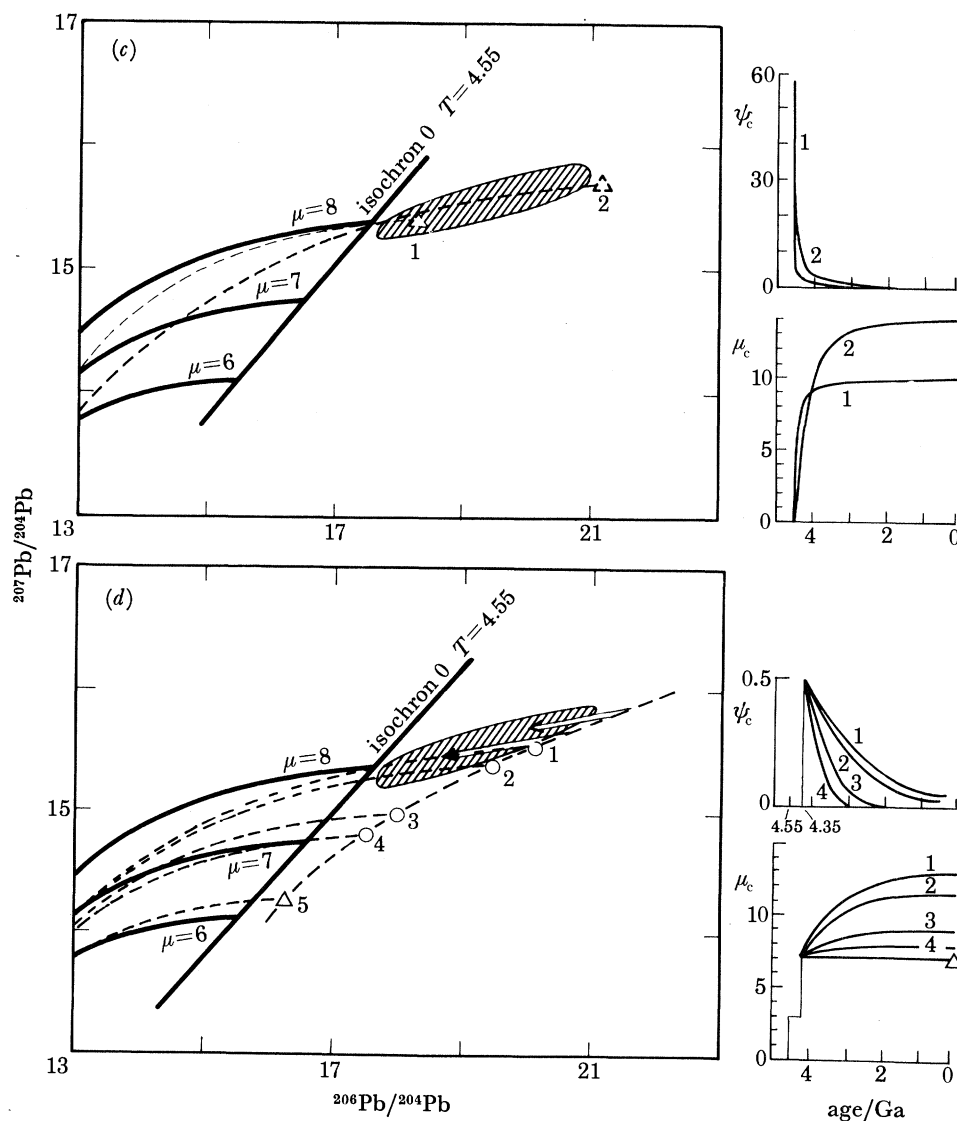


FIGURE 2. $^{207}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$ diagram showing the effects of U–Pb core fractionation on present lead isotopic composition. Closed system evolution curves and geochrons as well as the domain for m.o.r.b. are shown for reference (heavy line and hatched area respectively). For (a) we have assumed that fractionation, ψ_c , due to core formation starts either at 4.55 Ga (1) or at 4.35 Ga (2), with different intensities. For (b), ψ_c has been unequally efficient in different mantle domains 4.35 Ga ago but was subsequently similar everywhere (time constant 1.2 Ga). Points 1 to 3 thus represent domains with different initial values for μ . For (c), core fractionation starts at 4.55 Ga and rapidly increases in intensity. Points 1 and 2 correspond to different time constants. For (d), μ is homogeneous and equal to 3 until 4.35 Ga ago, when ψ_c was suddenly effective and decreased with different time constants in different mantle domains (from 0.3 Ga for point 4 to 1.2 Ga for point 1).

that the primary differentiation occurred when convection is on the scale of the whole mantle: the gravitational energy realized by such a process is so large that it is very difficult to imagine anything but a vigorous stirring of the whole mantle.

After such a 'hot episode', mantle convection breaks into two layers. At that time, the upper mantle already has a μ of 7 to 8, which does not vary too much because continental crust formation does not strongly fractionate U/Pb ratios. The growth curves for continental ore deposits give μ around 9.5 to 10.

The lower mantle continues to undergo exchange with the core, with diminishing intensity, and this exchange induces the extreme values for μ and lead isotope ratios observed for o.i.b.

Calculation of such a model also explains o.i.b. and m.o.r.b. data. The μ values for the lower mantle vary from about 7 to around 15 or 18.

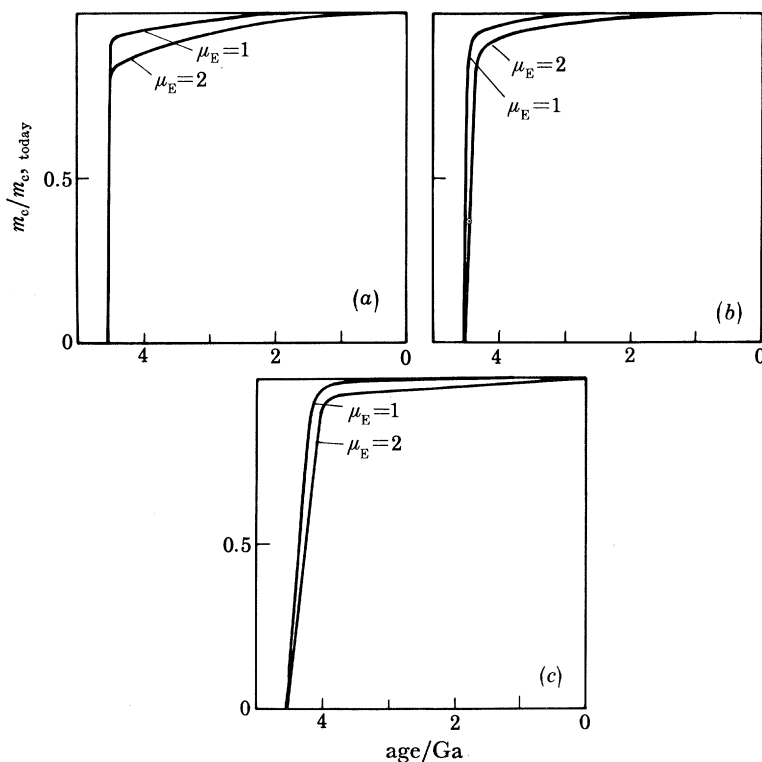


FIGURE 3. Growth in the mass of the core throughout geological time, according to various models and various values of $\mu_E = (^{238}\text{U}/^{204}\text{Pb})$ for the whole Earth. Cases (a), (b) and (c) correspond to those described in figure 2.

THE GROWTH CURVE OF THE CORE

From the calculated models we can infer a growth curve for the core according to several scenarios. Since the core contains no uranium, we may consider that $\mu_c = 0$. The chemical budget balance equation (neglecting the continental crust) can be written as

$$\mu_E = \mu_m(1 - a)$$

and

$$\mu_E = (^{238}\text{U}/^{204}\text{Pb})_{\text{bulk Earth}},$$

with $a = C_c m_c / C_E m_E$, $\mu_m = (^{238}\text{U}/^{204}\text{Pb})_{\text{mantle}}$, $C_c = ^{204}\text{Pb}$ concentration in the core, $C_E = ^{204}\text{Pb}$ concentration in the bulk Earth, $m_c =$ mass of the core, and $m_E =$ mass of the bulk Earth.

The budget equation can then be rewritten

$$m_c = m_E \left(\frac{\mu_m - \mu_E}{\mu_m} \right) \frac{C_E}{C_c}.$$

For translating the variation of $\mu_m(t)$ into $m_c(t)$ we need to know two parameters, C_E/C_c and μ_E , the latter having the largest uncertainty!

C_1 chondrites have a μ from 0.05 to 0.1 (Tatsumoto *et al.* 1973). However, Pb is a volatile element whereas U is a refractory one. Therefore, based on the fact that Rb/Sr for the Earth is much lower than Rb/Sr for C_1 meteorites we have to admit that the (U/Pb) ratio for the bulk Earth is greater than that for C_1 meteorites. By analogy with what we know about the Rb/Sr ratio, we think that a good estimate of μ_E is 1 or 2.

If we end up with $\mu_m = 15$ to 20, the m_c/m_t ratio today is 0.3252 and then we can deduce that the order of magnitude of $C_E/C_c = 0.35$.

Before doing any further calculations let us comment on these figures. Taking the values of lead concentration in troilite of iron meteorites to be about 7000 ng g⁻¹ (Tatsumoto *et al.* 1973), and putting something like 10% FeS into the core, $C_c = 700$ ng g⁻¹. In this case, $C_E = 250$ ng g⁻¹. Such a value is not ridiculous if we remember that C_1 chondrites contain 1500 ng g⁻¹ and metamorphosed chondrites 250 ng g⁻¹ (Tatsumoto *et al.* 1973).

Using these values, we may then compute a growth curve for the core throughout geological time (figure 3). Of course, such a calculation is different if we use the two-layer model, because we then have the ratio $m_c/m_{1,m}$, where $m_{1,m}$ is the mass of the lower mantle.

We may thus compute the core growth curve for the iron concentration and the Ni/Fe ratio of the mantle. From the chemical budget balance equation we get

$$C_{\text{Fe}, m} = \frac{C_{\text{Fe}, E} - C_{\text{Fe}, c} m_c/m_E}{1 - m_c/m_E}.$$

In the same manner,

$$\left(\frac{\text{Ni}}{\text{Fe}} \right)_m = \frac{\left(\frac{\text{Ni}}{\text{Fe}} \right)_E - \left(\frac{\text{Ni}}{\text{Fe}} \right)_c \frac{C_{\text{Fe}, c} m_c}{C_{\text{Fe}, E} m_E}}{1 - \frac{C_{\text{Fe}, c} m_c}{C_{\text{Fe}, E} m_E}}.$$

CONCENTRATION OF IRON IN THE MANTLE THROUGHOUT GEOLOGICAL TIME

Numerical values for insertion into these equations can be inferred from the mantle and core compositions and from the known values of the volume proportions between mantle and core. These values are given in the legend to figure 4; the m_c/m_E ratios can be found in figure 3.

It is worth noting that over 'geological' periods (i.e. between 3.5 Ga and the present) the iron content of the mantle (taken as homogeneous for simplicity) varies from 6.5 to 5.5% (in Fe, not FeO) for $\mu_E = 1$ and from 7.5 to 5.5% for $\mu_E = 2$ in model (a). In model (c), for $\mu_E = 1$ there is no variation of the iron content, whereas for $\mu_E = 2$ there is a variation of 6 to 5.5%.

The comparison of iron content estimates in the Archaean mantle by Duke (1979), who found a value of 7%, with that at present, 5.5%, tends to corroborate this model.

The Ni/Fe ratio variations are even more obvious. Within the same time span, in model (a) Ni/Fe varies from 0.09 to 0.04 for $\mu_E = 1$ and from 0.12 to 0.04 for $\mu_E = 2$. In model (c), where

iron variations are smaller, the Ni/Fe ratio variations remain significant: for $\mu_E = 1$, Ni/Fe varies from 0.055 to 0.04 and for $\mu_E = 2$ from 0.07 to 0.04.

If the core pumping model were valid, the time variations calculated should also exist for other siderophile elements.

A comparative study of siderophile element contents in ancient komatiites and in sub-recent peridotite should give a reliable test for the hypotheses.

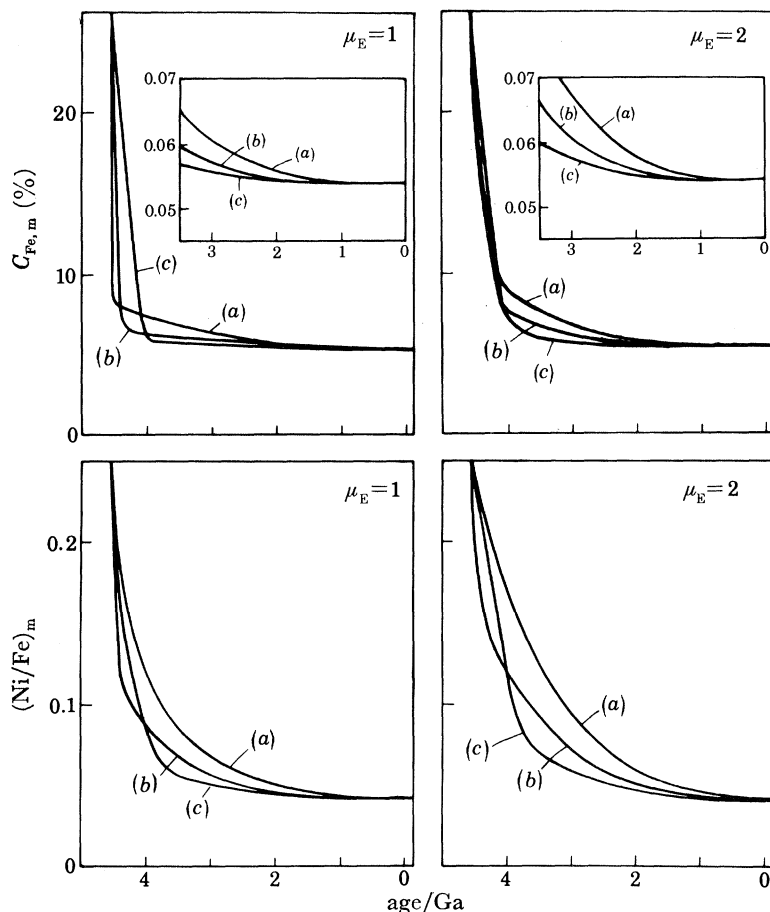


FIGURE 4. Evolution of Fe concentration and Ni/Fe ratio within the core throughout geological time, according to the models described in figure 2. Numerical values used for the calculations are: $C_{Fe,c} = 70\%$, $C_{Fe,E} = 26.4\%$, $(Ni/Fe)_c = \frac{2}{7}$ and $(Ni/Fe)_E = 0.252$.

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CHEMICAL ASPECTS OF CORE FORMATION

59

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