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THE CONTRIBUTION OF TRACE ELEMENT AND ISOTOPE GEOCHEMISTRY TO THE PETROGENESIS OF OCEANIC BASALTS AND THE COMPOSITION OF THE UPPER MANTLE

It is generally accepted by earth scientists that basalts are derived as silicate melts by partial melting of the Earth's upper mantle, and that seismological evidence and arguments based on temperature gradients suggest that primary basalt magmas originate at depths from tens to hundreds of kilometers. Because the oceanic crust is relatively thin the study of oceanic basalts has assumed particular importance because of the much greater chance that these basalts would be uncontaminated by crustal material, and would therefore afford one of our best links for studying the composition of the upper mantle. With sufficient knowledge of the way in which elements distribute themselves between these basaltic melts and various silicate minerals we can hope to distinguish between near-surface differentiation effects and those effects due to the partial melting process itself or related to the chemical and mineralogical make-up of particular regions of the mantle. Trace element and radiogenic isotope abundance studies have proved particularly useful in such studies; indeed the first suggestion that the upper mantle is chemically heterogeneous employed just such techniques [1, 2].

An important question relating to the intertwined problems of upper mantle composition and oceanic basalt genesis is to what extent the liquids produced by partial melting within the upper mantle are modified during transport to the surface. Further, if magmas do evolve before or during their ascent to the surface, is the principal mechanism that of:

(*a*) closed system eclogite fractionation as advocated by O'Hara [3] which predicts the formation of large volumes of high density eclogites within the upper mantle

or (*b*) closed system enstatite fractionation accompanied by extensive open system wall rock reactions as advocated by Green and Ringwood [4] which does not predict the formation of any accumulates in the upper mantle with densities much higher than peridotite?

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Initial constraints on the petrogenesis of oceanic basalts have been derived chiefly from experimental petrology, and have involved both experimental studies of artificial systems and natural basalts. Such an approach concentrates attention on bulk chemistry and mineralogy, and has, for example, led Green and Ringwood at Canberra to develop a uniform upper mantle source region for basaltic magmas consisting of a peridotitic composition (pyrolite) probably containing about 0.1 % to 0.4 % of water. They have attributed the presence of the low velocity zone (or asthenosphere) in the upper mantle to a small (∼ 5 %) degree of melting of pyrolite containing ∼ 0.1 % of water; the liquid fraction in the asthenosphere would then be a highly undersaturated olivine nephelinite. Other magma types of direct upper mantle derivation embracing tholeiites, olivine trachybasalts, tholeiitic picrites, etc., can on this model be assigned to a petrogenetic grid expressing the depth (pressure) of magma segregation and the degree of partial melting of the source pyrolite. Fig. 1. shows such a grid taken from a paper by Green [5]; it is largely self-explanatory, but requires the caveat that the magma types which appear on it are only those liquids developed in equilibrium with residual peridotite — these magmas may of course in turn be modified by crystal fractionation as they move from the

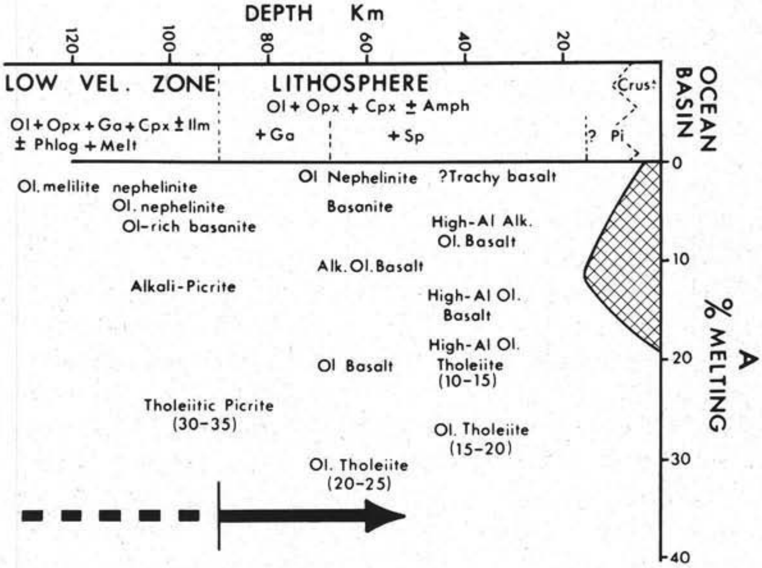


Fig. 1. — Diagram summarizing the petrogenetic applications of experimental melting studies on material of pyrolite composition. The column to the left illustrates the mineralogical character of the lithosphere and the depth to the onset of partial melting in the island arc situation and in the normal oceanic crust-mantle situation. In the right side of the diagrams, the character of magma derived by partial melting of pyrolite is plotted as a function of the depth of magma segregation and degree of partial melting. Numbers in parentheses adjacent to basalt names refer to the normative olivine content of the partial melt. The hatched areas illustrate the range of conditions over which the quartz-normative magmas may be derived by direct partial melting and magma segregation from pyrolite. The diagram is compiled for melting under water-undersaturated conditions with a water content of about 0.2 % in the source pyrolite.
[From Earth Planet Sci. letters *19* (1973), *51*].

depth of magma segregation to eruption at the Earth's surface. This genetic model can account quite well for variations in the major element chemistry and normative mineralogy among basalts.

It is time now to turn to the role played by the chemistry of the dispersed or incompatible elements K, Rb, Th, U, Ba and the rare earth elements (REE), and the restrictions on petrogenetic theories which they afford. In particular, can the basic Green and Ringwood theory based on a mantle of constant peridotitic (pyrolite) composition and single-stage melting, explain the incompatible element chemistry observed in oceanic basalts? The answer is no, but we should first review briefly the major features of this trace element chemistry, which is of such fundamental significance in the petrology of oceanic volcanics.

The classic paper in this field was that of Engel et al. in 1965 [6]. Recent oceanic studies had at that time shown that a distinctive tholeiitic basalt is the principal igneous rock encrusting the deeper parts of the oceans, distinguished by about 49-50 % silica, a low Fe_2O_3/FeO ratio and a high Na/K ratio. Engel et al. were able to show that the tholeiitic basalts that form most of the deeply submerged volcanic features, especially oceanic rises, in the oceans are characterised by extremely low contents of Ba, K, P, Pb, Sr, Th, U and Zr, by $Fe_2O_3/FeO < 0.2$ and $Na/K > 10$, and by $K/Rb \sim 1300$ and $^{87}Sr/^{86}Sr \sim 0.702$, similar to what was then known about calcium-rich basaltic achondrites. They took the close compositional similarities between oceanic ridge tholeiites and Ca-rich achondrites to indicate the relatively primitive nature of oceanic abyssal tholeiites.

In contrast they found the alkali-rich basalts that cap submarine and island volcanoes, even where these occur close to (but on the flanks) of ocean ridges, to be relatively enriched in Ba, K, La, Nb, P, Pb, Rb, Fe_2O_3, Sr, Ti, Th, U and Zr, i.e. in the same elements that are enriched in the sialic continental crusts by factors of 5 to 1000 more than the amounts inferred to be present in the upper mantle. These analytical data coupled with field relationships led Engel et al. to postulate that alkali-rich basalts are derivative rocks, fractionated from oceanic tholeiites by magmatic differentiation processes, and that oceanic tholeiites are the principal or only primary magmas generated in the upper mantle beneath the oceans. They further suggested that oceanic tholeiites are either complete melts of the upper mantle or are generated from a mix of this tholeiite with a magnesium rich peridotite, and so were led to the conclusion that oceanic tholeiites are the least contaminated and only common or partial melt of the upper mantle now available for study.

From a comparison of chemical compositions of island volcanics with those of abyssal tholeiites they were able to demonstrate systematic effects accompanying the progressive enrichment of alkalis in oceanic volcanic rocks, such as:-

K/Rb decreases from ~ 1300 in abyssal tholeiite to ~ 350 in alkali basalts;

Sr/Rb decreases from ~ 130 in abyssal tholeiite to as low as ~ 9 in alkali basalts;

K Zr increases from ∼ 14 in abyssal tholeiite to ∼ 50 in trachytic pumice. They interpreted these systematic changes as further evidence of the derivative origin of the alkali basalts from the parental oceanic tholeiites, or, in a 1971 restatement by Engel [7], derived either from a parental magma of tholeiitic composition, or from magmas parent to the tholeiites themselves.

The next major step forward in the delineation of the restrictions from dispersed element geochemistry to theories of the genetic relations between magma types came in the classic paper of Gast in 1968 [2]. This paper explored in some detail the partitioning of dispersed elements (elements that do not exist as a major stoichiometric component of any phase in the system under consideration) between liquid and solid phases due to crystallization and partial or complete melting under complete or surface equilibrium conditions, reviewed existing dispersed element concentration data in oceanic basalts, and outlined the restrictions on petrogenetic relationships which the data and model implied.

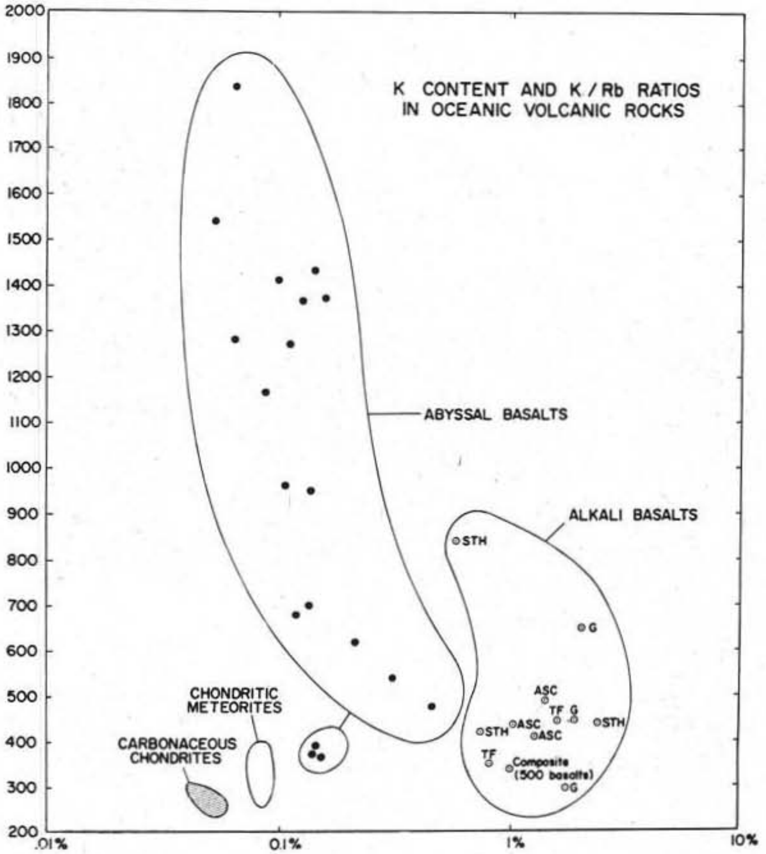


Fig. 2. — K/Rb ratios and K contents of some oceanic volcanic rocks. [From Geochim. et Cosmochim. Acta *32* (1968), 1057].

In summary, apart from a confirmation of the characteristic features noted by Engel and his co-workers, Gast noted, as shown in Fig. 2 that the abyssal basalts have high K/Rb ratios and very low K and Rb contents when compared with the alkaline basalts. Since Hart [8] has questioned the freshness of some of the abyssal basalts used by Gast, it is worth while to note Fig. 3 which gives modern abyssal basalt data due to Hart and others, O'Nions and Pankhurst, as well as several modern oceanic alkali basalt analyses. It will be seen that the modern data also shows a clear difference between abyssal tholeiites and island volcanics, but *does not support for the abyssal basalts a trend of decreasing K/Rb with increasing K as proposed by Gast and reiterated by Kay, Hubbard and Gast* [9]. Another important feature is brought out by Fig. 4 which shows that the alkaline basalts are much enriched in light rare earths relative to heavy rare earths when compared with

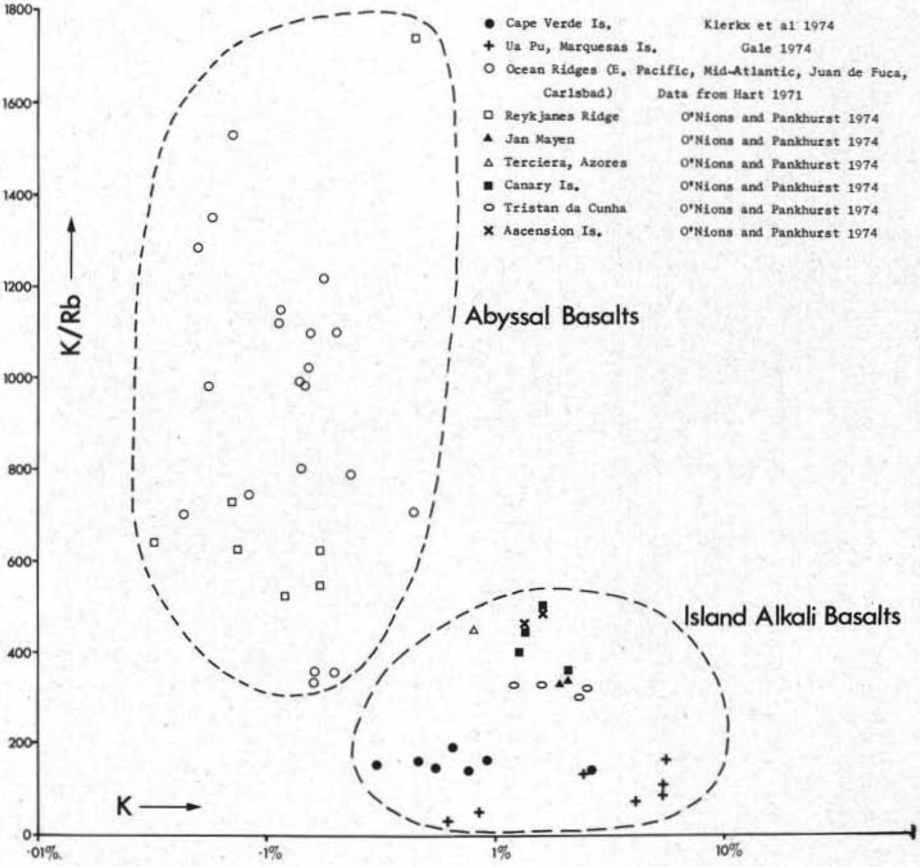


Fig. 3. — Modern data on K/Rb ratios and K contents of abyssal and oceanic island rocks.

abyssal basalts. Extensive more recent work, especially by Schilling [10], has confirmed this feature, and demonstrates a remarkable rare earth uniformity of mid-ocean ridge tholeiites which in itself seems to require a unique and simple volcanic process to be operative. Schilling believes that it calls for upward migration of melt and slush from a relatively homogeneous source in the mantle — the asthenosphere, or low velocity layer — probably followed by further partial melting during ascent. A further feature is that the barium content of abyssal basalts is much more depleted than is strontium relative to alkali basalts.

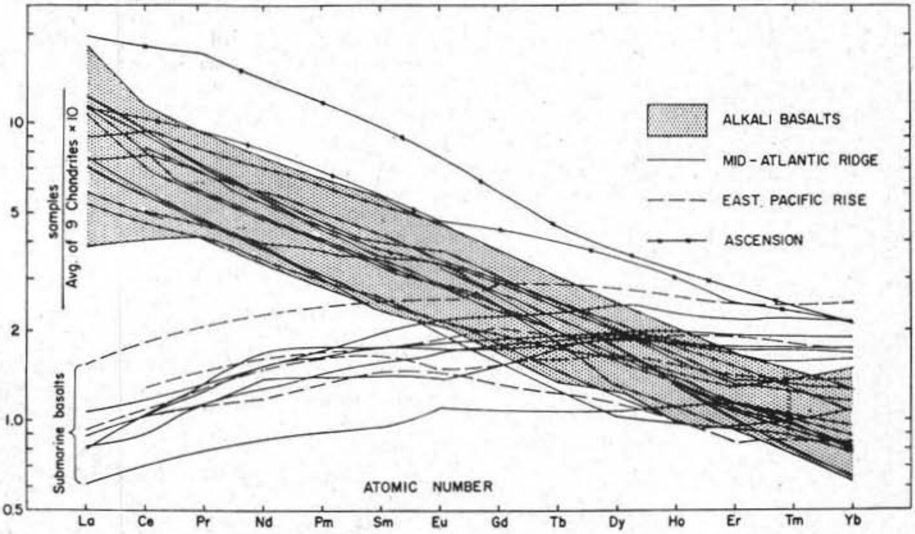


Fig. 4. — The relative abundance of rare earth elements in abyssal basalts and alkali basalts from oceanic islands.
[From Geochim. et Cosmochim. Acta *32* (1968), 1057].

It is appropriate now to turn to a brief consideration of the ways in which Gast was able to constrain petrogenetic theories on the basis of the observed incompatible element concentrations, the observed trace element distribution coefficients and a consideration of Rayleigh surface equilibrium crystal formation, or partial melting, models. First, what of the idea advocated by Engel and others that alkali basalts are derived from a sub-alkaline or tholeiitic liquid? Gast was able to show that it is extremely difficult to enrich Ba, K and Rb by factors of 10 or more by crystal fractionation without crystallising 90% or more of the parent liquid. Any appeal to volatile transfer as an alternative mechanism for enrichment of alkalis in alkali olivine basalt meets the difficulty that it is exceedingly unlikely that such a process should also enrich such dissimilar elements as Rb, Ba, U, Th, La and Pb to a similar degree. Again, elements such as K, Rb, Ba, U and Pb that are highly enriched in a residual liquid cannot be readily fractionated from each other by fractional crystallization, so that this mechanism is highly unlikely to be competent to derive alkali basalts with K/Rb ratios of 150 to 500 from parent liquids with K/Rb ratios of 600-1800. Thus the idea that alkali basalts are derived from parental tholeiitic magmas, or that these two basalt types are genetically related in any way by fractional crystallization, is unlikely on the basis of a proper consideration of the incompatible trace element data.

Gast also made extensive calculation of batch or partial melting and fractional (continuous accumulation) melting of three mineral assemblages that may occur in the upper mantle:-

Lherzolite (clinopyroxene, orthopyroxene, olivine and spinel);
Garnet-peridotite (clinopyroxene, garnet, orthopyroxene and olivine);
Amphibolite (horneblende, orthopyroxene, olivine and spinel);

together with consideration of the effect of trace phlogopite. One important general conclusion was that the incompatible trace element chemistry and the major element chemistry of liquids produced by partial melting are in many cases rather effectively decoupled, with the result that different mechanisms of production of the same bulk chemistry may result in quite different trace element compositions — thus illustrating the power of incompatible element chemistry in discriminating different petrogenetic mechanisms.

Gast's calculations were made with the aid of the assumption that a liquid created in the mantle is in approximate thermodynamic equilibrium with its coexisting phases when formed and with a collection of estimated distribution coefficients describing this equilibrium. Detailed consideration of the fractionation of the element pairs La/Yb, K/Rb, Ba/Sr and Cr/Ni showed that their absolute and relative abundances in alkaline magmas are entirely consistent with the hypothesis that these liquids are produced by small amounts (3-7 %) of partial melting of these accepted upper mantle mineral assemblages, i.e. of a more primitive trace element rich source. The element pair K/Rb forcs the mantle source of alkali basalts to have a K content of .03 to .04 % and K/Rb ratios of 500 to 1000.

It was also found to be a consequence of the model that the residues produced by these small amounts of partial or fractional melting would have Ba depleted relative to Sr, La and Ce relative to heavy REE and Rb depleted relative to K, whilst a variety of large ions would be depleted in these residues. These are distinguishing characteristics also for abyssal tholeiites, and when this information is combined with the result of the calculations that extensive partial melts of $\sim$ 15 % of the assemblages considered will result in liquids in which the dispersed element ratios are only slightly fractionated relative to the bulk solids, a mechanism for generating the abyssal basalts is suggested. The postulate is that these basalts are produced by extensive (15-40 %) melting of upper mantle regions that have been chemically fractionated by previous much less extensive partial melting events.

Many people (e.g. O'Hara, Green and Ringwood, Kushiro, Kuno) have argued that abyssal basalts and island basalts are genetically unrelated. It is probably generally accepted by most people nowadays that the weight of trace element data favours Gast's interpretation in general terms, requiring variable source composition as well as variations in conditions of melting, and it seems certain that abyssal and alkaline basalts cannot be genetically related by fractional crystallisation, and that the trace element data also rules out any interpretation of the Hawaiian volcanic petrology in terms of crystal fractionation of parental olivine tholeiite magmas.

An alternative suggestion of a mode of enrichment of the incompatible elements in a fractionating magma was the « wall rock » reaction of Green and Ringwood [4], which was envisaged as a highly selective contamination of a magma by its wall-rock environment involving complementary processes of reaction and extraction of the lowest melting fractions from the wall rock. This concept was severely and cogently criticised by Gast on grounds of the physical and chemical inadequacies of the mechanism as originally proposed, but has more recently been defended by Green [11] by a consideration of the role which water might play. Briefly the argument is that if a body of magma, undersaturated in water, moves through a mantle region containing hydrous phases, these phases may become unstable, their breakdown producing water and other volatiles which move into the magma and carry incompatible elements with them. Elements formerly substituting in amphibole or phlogopite but incompatible with pyroxenes and olivine could in principle move in this way. It will be interesting later on to see what modern isotopic studies have to say on this question.

To round off this introductory part of the talk, before moving on to a consideration of the further information given by isotopic studies, we may attempt to set the conclusions from trace element studies in the context of geophysical ideas. First we observe that if basalts with marked differences in incompatible elements, but with similar major element composition, were randomly distributed in space and time, then one might conceive of a mantle source region which is irregularly and randomly inhomogeneous due to effects of variable degrees of magma extraction at various times in the past. However, the regular appearance of the low K tholeiite with monotonously similar rare earth patterns [10] at mid ocean ridge crests suggests that this magmatic process taps a widespread and relatively homogeneous source region of the mantle. Basalts with high contents of incompatible elements do occur at abyssal depths in the ocean basins, but commonly on the flanks of ridges and in seamounts which locally are symmetrically developed on either side of the median valley or from chains moving away from the ridge crest in the direction of oceanic plate movements. These occurrences show that an oceanic crustal locality, which taps a source of low K tholeiite when at the ridge crest, may later tap a second source region when it moves away from the ridge crest; this second source characteristically provides magmas enriched in incompatible elements.

These facts may easily be reconciled with the thermal model of Oxburgh and Turcotte [12] in which they define a « zone of partial melting » in a convecting mantle. This zone is defined in terms of the difference between a predicted model temperature and the olivine tholeiite fusion temperature. Fig. 5 reproduces their predicted zones and material flow lines in the region of an ocean ridge. If one traces the path of a unit volume of mantle taken progressively further away from the axis of the ascending plume, it is seen that the depth where this unit volume experiences its greatest excess temperature (increment above the fusion temperature) increases from a minimum depth of approximately 50 km to depths of more than 100 km. Moreover, the magnitude of the « excess temperature » that is experienced also decreases in going away from the axis. If one postulates also that the degree of partial melting that results from a given temperature excess will decrease with an increase in pressure, than it seems likely that the extent of partial melting away from the central plume will be much less than that in the shallow region over the rising plume. Both the chemical properties and the spatial distribution of oceanic volcanic materials are broadly in accord with this now rather old model.

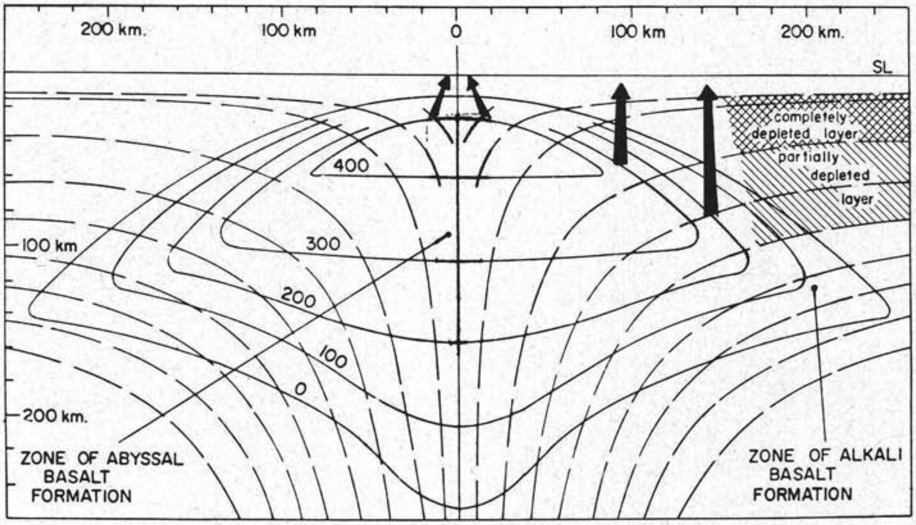


Fig. 5. — Oxburgh and Turcotte model of a mantle convection cell showing possible sites of alkali basalt and abyssal basalt generation. Note that abyssal basalt liquid cannot become enriched in the dispersed elements during passage through surrounding solid matter, since this solid matter will be depleted in those elements enriched in a liquid from previous partial melting. In contrast the alkali basalt liquids traverse mantle regions that may have been only partially depleted, and so the alkali basalt liquids may be enriched in dispersed elements during their vertical transport.
[Taken from Geochim. et Cosmochim. Acta *32* (1968), 1057].

Against this background, what further information is provided from studies of lead and strontium isotopes?

Isotopic Measurements in Oceanic Volcanics

A large number of Sr and Pb isotopic measurements have been reported from many oceanic islands; recent reviews of the Sr data have been given by Faure and Powell (1972) in their monograph on strontium isotope geology, of the Pb data by Oversby and Gast [13] and of both Sr and Pb data in the paper by O'Nions and Pankhurst [14]. For Sr the $^{87}Sr/^{86}Sr$ ratios show very considerable scatter, both overall (0.702 to 0.706) and in some cases within a single island; such a variation might be thought to be of considerable petrogenetic significance. However both the Sr data and the Pb data have in the past been subject to considerable analytical uncertainties, and there has been far too little control of either Pb or Sr analyses with reference to properly certified standards, so that the quality of most previous data has been inadequate to demonstrate true consistency or small differences.

Over the past few years there has been a revolution in the technical aspects of Pb and Sr isotope geology. Highly precise certified isotopic standards for Sr and Pb are now available from the U.S. National Bureau of Standards, so that confident intercomparison between work from different laboratories is now possible, and chemical separative techniques have been developed which allow the blanks for both Sr and Pb to be pushed down from the hundreds of nanograms level to the sub-nanogram level. Following the pioneering work of Papanastassiou and Wasserburg at the California Institute of Technology, a number of laboratories have now proved their capability of measuring $^{87}Sr/^{86}Sr$ ratios to a relative precision of about 0.005 % (i.e. 1 part in 20,000), compared with an average of not better than 1 part in 7000 some years ago. The independent discovery by the Russians and the Americans of the silica gel activator method for the mass spectroscopy of lead, and its subsequent refinement, have made possible measurements of $^{207}Pb/^{204}Pb$ ratios etc. to an accuracy of better than 0.1 %, whereas the PbS method formerly commonly used was about an order of magnitude less accurate. For both Sr and Pb it is now easily possible to make precise isotopic abundance measurements on less than 100 nanogram samples. With the advent of these improvements there is now a clear need to make extensive new isotopic measurements of oceanic volcanics. There is already quite a body of precise new data for Sr, chiefly from laboratories in the Carnegie Institution, Brussels and Oxford, whilst Tatsumoto and Sun have much unpublished new Pb isotope data. Let us now try to see what further constraints on petrogenesis the isotopic data affords, both with respect to earlier theories of the relationship between ocean island and ocean ridge basalts and with respect to the deep mantle plume hypothesis introduced recently by Morgan [27].

I shall attempt to sketch out the information provided by isotopic evidence in roughly the historical order of the development of this topic. The isotopic composition of Sr was used by Gast as early as 1960 [15] as strong evidence that the earth is chemically distinct from chondrites. The present-day $^{87}Sr/^{86}Sr$ ratio of the mantle can be roughly inferred from that found in oceanic volcanic rocks; from this ratio and the presumed initial ratio for the whole earth, one can infer the Rb/Sr ratio of the upper mantle source (assuming single-stage growth of radiogenic Sr) from the relation:

$$\frac{^{87}\mathrm{Sr}}{^{86}\mathrm{Sr}}\ \mathrm{source} = \frac{^{87}\mathrm{Sr}}{^{86}\mathrm{Sr}}\ \mathrm{initial} + \frac{^{87}\mathrm{Rb}}{^{86}\mathrm{Sr}}\ \mathrm{source} \times [\exp \lambda t - 1]$$

Inserting the best guesses of $(^{87}\mathrm{Sr}/^{86}\mathrm{Sr})_{\mathrm{INITIAL}} = 0.6989$ (the BABI value from basaltic achondrite meteorites) and $(^{87}\mathrm{Sr}/^{86}\mathrm{Sr})_{\mathrm{SOURCE}} = 0.70280$ (the best estimate from modern abyssal tholeiite data) and the presumed 4.5×10^9y age of the mantle, then (Rb/Sr) $\sim$ 0.02 for the present mantle, which compares with 0.1 to 0.8 for chondritic meteorites. We conclude that the Rb/Sr ratio of the Earth is about one-tenth that of the chondritic meteorites and the primitive solar nebula. Perhaps more important for our present purposes is the fact that the computed mantle source Rb/Sr ratio of $\sim$ .02 is up to several times greater than actually observed in modern analyses of some ocean ridge tholeiites, and since Rb is expected to partition more readily than Sr into the liquid phase upon partial melting, the mantle source of these tholeiites must have had lower Rb/Sr ratios still than the tholeiites. Thus the ridge tholeiite $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$ ratios (which modern analyses put in the range .7025 to .7030) are not supported by their present-day mantle source (Rb/Sr) ratios, and it follows that the mantle sources of ocean ridge tholeiites must have undergone depletion of Rb relative to Sr at some stage prior to magma formation. This is in accordance with the observation that these source regions are also relatively depleted in other incompatible elements.

It was realised quite early on that there were considerable differences in $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$ within the ocean basins, with this ratio varying quite widely from island to island, and since the decay rate of Rb is low and the Rb/Sr ratio is low it was argued that long periods of time are implied by the isotopic variations. Lead isotope measurements were also interpreted to suggest that mantle inhomogeneities had lasted for of the order of 1 or 2 thousand million years, notwithstanding the difficulty of understanding this in terms of a convecting mantle. Such discussion centre around the plot, for oceanic basalts, of $^{207}\mathrm{Pb}/^{204}\mathrm{Pb}$ against $^{206}\mathrm{Pb}/^{204}\mathrm{Pb}$, which is shown in Fig. 6. In this diagram single-stage closed system evolution for the mantle would result in the source regions falling along an isochron (the Geochron) with slope corresponding to 4.5×10^9y and passing through the composition of primordial lead. The exact position on this line would depend only on the U/Pb ratio for the system, more usually referred to in terms of μ-value ($= {}^{238}\mathrm{U}/^{204}\mathrm{Pb}$). Fig. 6 shows that many published Pb compositions do fall close to this line, including data for ocean-ridge basalts, for mainland Iceland, and for volcanic rocks from Tristan da Cunha, as well as many of the analysed basalts from Pacific Ocean islands. However, the scatter shown by this data is well outside analytical error, and requires a multi-stage evolution. Moreover, data for the Canary Is., Ascension Is., and St. Helena plots far to the right of the Geochron, requiring that the Pb developed for a considerable time in an environment with higher μ-values than for the ridge tholeiites. On the assumption of *equilibrium* melting, so that the Pb isotope ratios ore identical with those in the source regions (even though the U and Pb concentrations may not be), the extreme variation in lead isotope ratios requires fractionation events occurring sufficiently long ago for these differences to have been caused by radioactive decay. The approximately linear trend of the data in Fig. 6 has been taken to indicate a major fractionation of U from Pb in the mantle about 1000 to 2000 my ago (e.g. Ulrych 16), whilst more precise linear trends for individual islands have been taken to indicate 3-stage U/Pb fractionation, e.g. for the source of the S. Helena volcanics at 4.1×19^9y and 1.8×10^9y [17]. A further conclusion was that the depleted source of the ridge tholeiites is characterised by low U and low U/Pb ratios, and that these are relatively much higher in the source regions of alkali basalts.

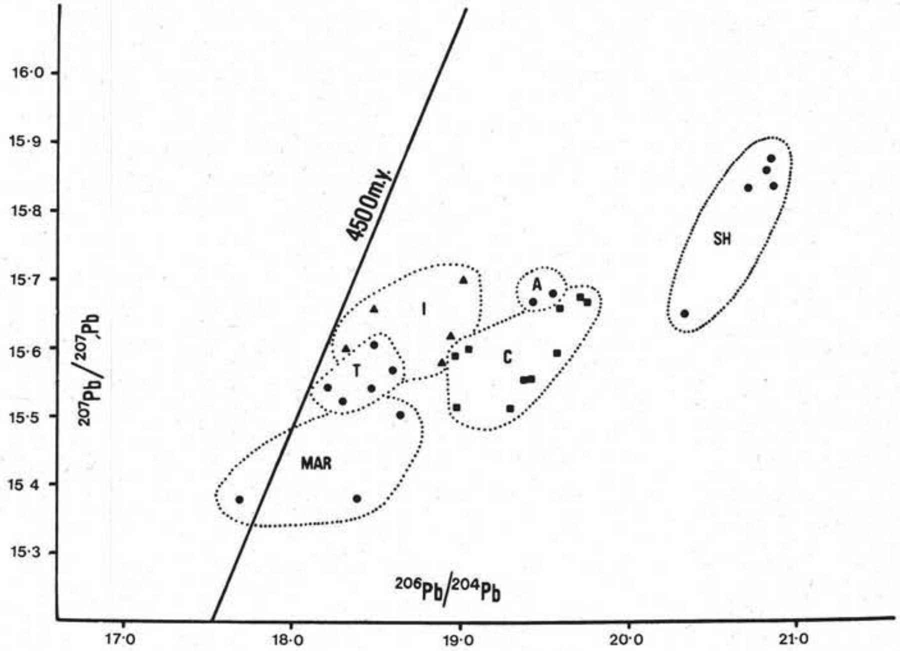


Fig. 6. — Pb-isotope plot for basaltic rocks from Mid-Atlantic Ridge islands. The 4500 my isochron is drawn through the least radiogenic Canyon Diablo meteorite lead. A = Ascension, C = Canaries, I = Neovolcanic zones of Iceland, MAR = Reykjanes Ridge, SH = St. Helena, T = Tristan da Cunha. [From J. Petrol. *15* (1974), 603].

I want now to turn to a consideration of more recent high-precision Sr-isotope data. This data has come chiefly from the following laboratories: Hedge and Peterman at the U.S.G.S. in Denver, Hart and his collaborators at the Carnegie

Institution, O'Nions and Pankhurst at Oxford, and, for unusual suites of alkalic rocks from the Cape Verde Is. in the Atlantic and for the Marquesas Is. in the Pacific, from Deutsch and her collaborators in Brussels and myself in Oxford respectively. It follows the pioneer work of Tatsumoto et al. in 1965 on ocean ridge tholeiites [18] which was followed by Gast in 1967 [19] and Ferrara et al. in 1969 [20] in their work on Eastern Pacific dredge samples.

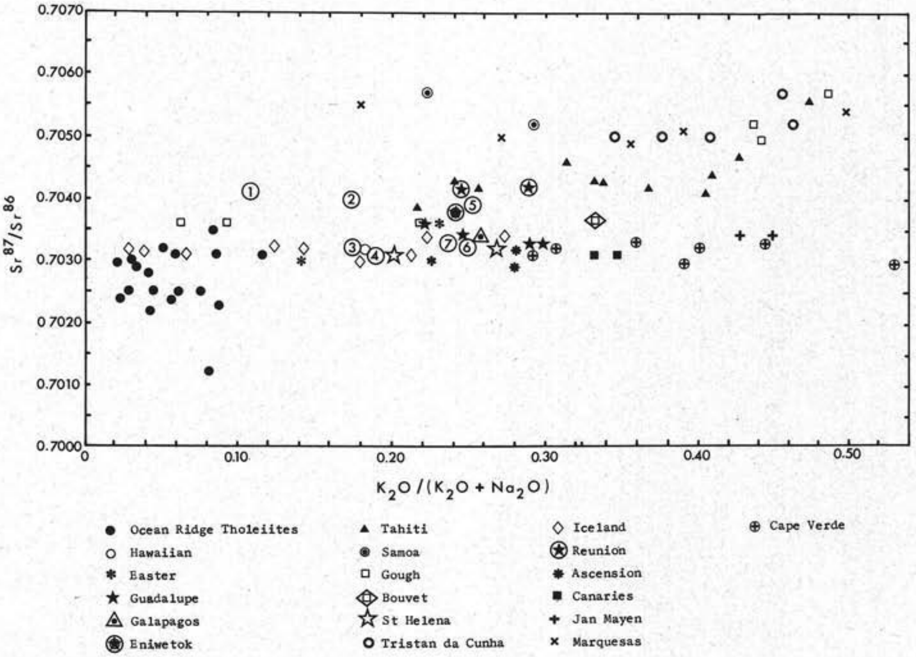
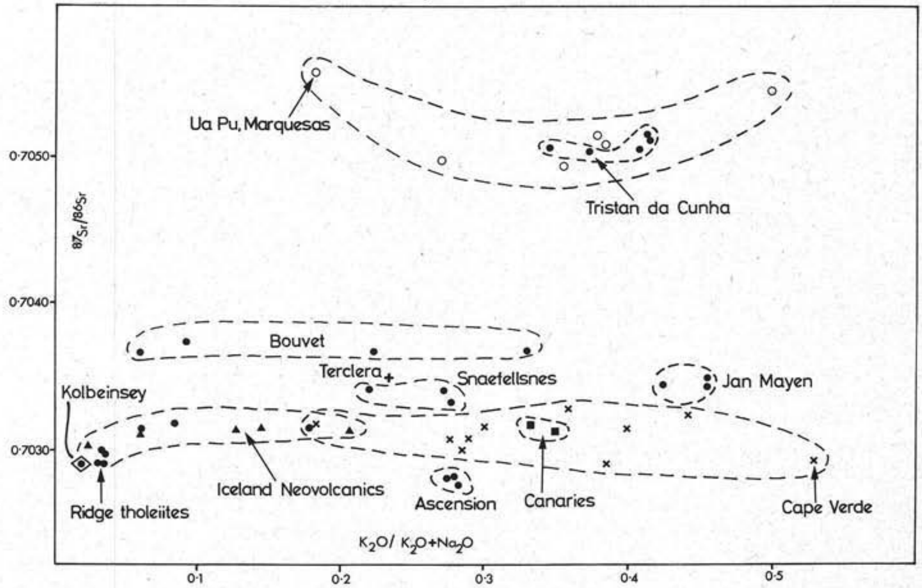


Fig. 7. — $^{87}Sr/^{86}Sr$ initial ratios plotted against $K_2O/(K_2O + Na_2O)$ for oceanic ridges and islands, including data plotted by Peterman and Hedge in Bull. Geol. Soc. Amer. *82* (1971), 355 together with more modern data presented in references 14, 22 and 23.

The most comprehensive recent set of Sr isotope data comes from the work of O'Nions and Pankhurst [14], and is for the Atlantic Ocean. The major islands included in their study extend along the length of the Mid-Atlantic Ridge from Bouvet Is. to Jan Mayen, whilst their study includes samples from the Reykjanes Ridge, the Charlie Gibbs Fracture Zone, and the island of Kolbeinsey to the north of Iceland.

I shall first of all use the totality of new data to dispose of a myth. Peterman and Hedge, in searching for a sympathetic variation between $^{87}Sr/^{86}Sr$ and other geochemical parameters, claim in their 1971 paper [21] to have demonstrated a positive correlation between $^{87}Sr/^{86}Sr$ and the ratio $K_2O/(K_2O + Na_2O)$ for basalts from the Atlantic and Pacific Oceans; their evidence for this is given in their

Fig. 1., which includes much older data. The correlation is not too convincing even there, but Fig. 7 shows the effect of including the new data and shows that the correlation, which depended chiefly on alkali basalts from oceanic islands, is essentially destroyed. If we restrict consideration only to the most modern data from O'Nions and Pankhurst [15], Klerkx, Deutsch and Paepe [22], and Gale [23], then Fig. 8 shows that this data does not support a correlation. It is necessary, therefore, to reject the conclusion of Peterman and Hedge that such a correlation exists, is of fundamental significance, and that the higher the alkali content of an erupted alkali basalt, the closer its source region approximates to primitive mantle which has retained its primordial alkali content and Rb/Sr ratio. Instead the most striking conclusions to be drawn from the plot of $^{87}Sr/^{86}Sr$ against $K_2O/(K_2O + Na_2O)$ are that within an individual island there is generally a complete lack of correlation, and that even for the very strongly alkalic basalts of the Cape Verde Is. and Ua Pu the isotopic ratio remains essentially constant with increasing K-content.



Post 1973 Data

Cape Verde - Klerkx, Duetsch and Paepe; Ua Pu - Gale; All other data due to O'Nions and Pankhurst.

Fig. 8. — $^{87}Sr/^{86}Sr$ initial ratios plotted against $K_2O/(K_2O + Na_2O)$ for Mid-Atlantic Ridge and oceanic island rocks; modern data only. Cape Verde data from reference 22, Ua Pu from reference 23, all other data from reference 14.

On the other hand the diagram shows that there is a clear distinction to be drawn between abyssal tholeiites and island alkali basalts, with the abyssal tholeiites being characterised not only by low incompatible element contents but also by low ^{87}Sr/^{86}Sr ratios. We can also see that the source region of the Ascension lavas has been, in terms of time-averaged Rb/Sr and ^{87}Sr/^{86}Sr ratios, indistinguishable from that of abyssal tholeiites for the greater part of the Earth's history, despite the great dissimilarity of the volcanic products.

Let us review the chief findings of the modern Sr data as summarised in Fig. 8.

(*a*) *Abyssal ocean ridge tholeiites* from the Reykjanes Ridge from just south of Iceland to as far as the Charlie Gibbs Fracture Zone, and from Kolbeinsey Is. north of Iceland, give essentially the same ^{87}Sr/^{86}Sr ratio of 0.70294 ± 4.

(*b*) *Recent ocean island tholeiites* (from the Neovolcanic Zones of Iceland and from the Canaries) show a very limited range of ^{87}Sr/^{86}Sr ratios about a mean of 0.70313, except that for Bouvet in the extreme south Atlantic tholeiites and trachytes have the identical value of 0.70369 ± 3.

(*c*) *Alkali ocean island basalts* yield a wide range of ^{87}Sr/^{86}Sr ratios, with a predominant tendency to values higher than for abyssal tholeiites, going up to 0.70504 ± 6 for Tristan da Cunha, and even up to 0.70556 for strongly alkalic basalts from Ua Pu, Marquesas Is., in the Pacific. However, the alkali-olivine basalts from Ascension Is. give values about 0.70280, slightly lower even than for the Reykjanes Ridge.

(*d*) *^{87}Sr/^{86}Sr ratios within a single island* tend to be constant, with intermediate compositions (trachybasalt, trachyandesite and trachyte) often having ratios indistinguishable from their associated basalts. Where this is true it argues for simple petrogenetic relationships between the various magmas, and against significant amounts of crustal contamination. In fact the Sr-isotope evidence seems to be strong enough to justify the statement that in general the long-standing ghost of crustal contamination of oceanic volcanics is finally laid.

An obvious exception to this rule is Iceland, which is much larger, has a longer volcanic history and has a more complex crustal structure than any of the other islands. Here, as shown in Fig. 9. there has been a secular decrease in the ^{87}Sr/^{86}Sr ratio of erupted tholeiites from 0.70346 about 16 6m.y. ago to 0.70313 in Recent times; as O'Nions and Pankhurst [17] have said, this relationship is compatible with (*a*) melting different parts of an inhomogeneous source, (*b*) a mantle plume hypothesis, (*c*) mixing hypotheses or (*d*) disequilibrium melting of an homogeneous source. I shall return later to a consideration of the latter two topics.

The results of this modern work demonstrate three features which most clearly require explanation, and which must have the greatest petrogenetic significance:

1. The clear difference between ridge tholeiites and island basalts.
2. The generally small variation of ^{87}Sr/^{86}Sr ratio between the alkali basalts and their derivatives on any one island.

3. The considerable variation in $^{87}Sr/^{86}Sr$ *between* islands along the Mid-Atlantic Ridge, or in the Pacific Basin.

Further features of the Sr data are brought out by an isochron plot, as in Fig. 10; it will be noted that the island alkali basalts fall on, or to the right of, the 4.5×10^9yr. isochron. This clearly can be taken to illustrate the expectation that, in a small degree of partial melting such as is represented by alkali basalt magma, the Rb/Sr ratio of the liquid is considerably increased over that of the source region. This is obvious in the case of the Ascension lavas, which have Rb/Sr ratios and trace element contents comparable with the Tristan lavas, but $^{87}Sr/^{86}Sr$ ratios which are so low that they indicate a small partial melt of a source region which has previously experienced a Rb/Sr evolution almost identical to that of ocean ridge tholeiites for the greater part of the Earth's history.

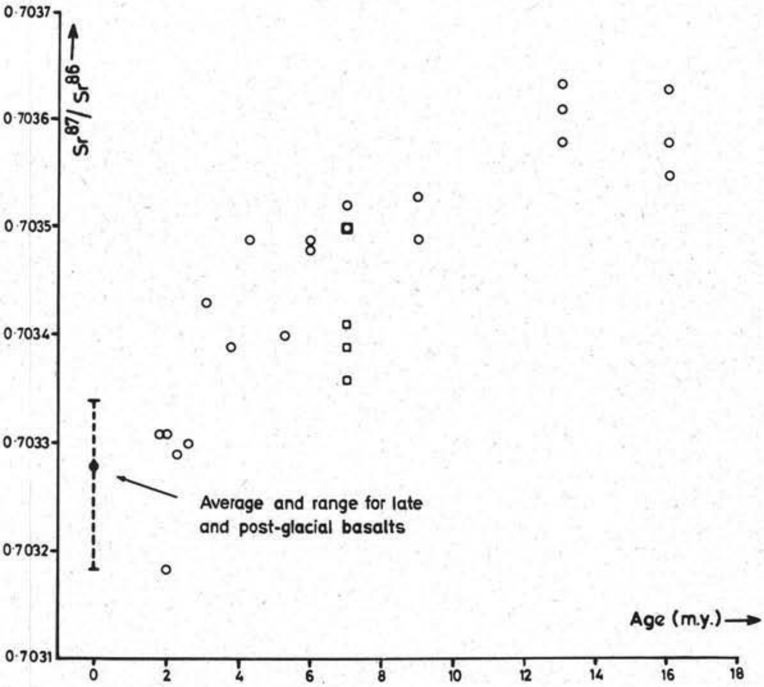


Fig. 9. — Variation of initial $^{87}Sr/^{86}Sr$ with time for Tertiary and Pleistocene igneous rocks from Iceland. Circles are for basic rocks and squares are for acid rocks.
[Taken from Earth Planet. Sci. Letters *21* (1973), 13].

I have already said that there is nothing to support the contention of Peterman and Hedge that alkali-basalts represent partial melts of a primitive mantle. About the only really definite conclusion to be made at present from the Sr isotope data is that the source regions of alkali basalts have generally been associated with higher Rb/Sr ratios, time-averaged over the past 4.5×10^9yrs., than those of the ocean ridge tholeiites. Moreover, the wide variations in ^{87}Sr/^{86}Sr of oceanic basalts may be taken to provide direct evidence for inhomogeneous distribution of Sr isotopes in their mantle source region, provided that we assume that the basalts were produced by equilibrium melting. It is clearly very important to know on what scale such heterogeneities exist — on the mineral scale or in domains of much larger extent? — and also over what periods of time they have developed.

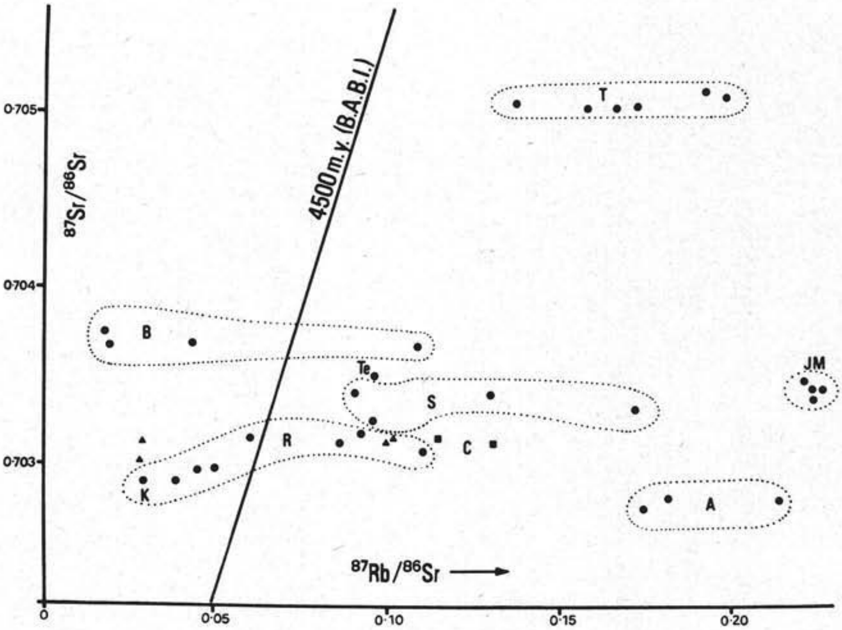


Fig. 10. — ^{87}Sr/^{86}Sr plotted against ^{87}Rb/^{86}Sr for oceanic basalts. The isochron corresponds to single stage systems which had initial ^{87}Sr/^{86}Sr ratios equal to those of basaltic achondrites 4500 my ago. A = Ascension, B = Bouvet, C = Canaries, JM = Jan Mayern, I (triangles) = Iceland (Neovolcanics), K = Kolbeinsey, R = Reykjanes Ridge and Charlie Gibbs Fracture Zone, S = Snaefellsnes, T = Tristanda Cunha, Te = Terciera.
[Taken from J. Petrol. *15* (1974), 603].

Equilibrium Melting

In most discussions of trace element distributions or of Sr and Pb isotope variations in oceanic basalts it has been assumed that, at the high temperatures required for partial melting of their source material (peridotite?), reaction and diffusion kinetics would be so fast as to ensure bulk equilibrium between the liquid and the solid residual phases, so that isotopes would be completely homogenized. This assumption lies at the base of Gast's 1968 [2] hypothesis that alkali basalts are derived by a few per cent of melting from a primitive mantle, whilst the ridge tholeiites are derived by 15-40 % melting of the depleted residue.

However, the isotopic work places a further constraint on this trace-element derived model, in that the widely different Sr and Pb isotopic compositions found do not allow *current* derivation of the two types of oceanic basalt, in this way, from a uniform mantle. The fractionation events must have occurred sufficiently long ago for these differences to have been generated by radioactive decay. If we use the measured Rb/Sr for specific island basalts as an upper limit for their immediate source region, then we can compute the time required for the $^{87}Sr/^{86}Sr$ ratios of specific island basalts to develop in isolation from the $^{87}Sr/^{86}Sr$ value appropriate to the source regions of basalts erupted on the ridge itself; such estimates range from 200 m.y. to several thousand million years for different islands.

Thus the inevitable consequence of the equilibrium melting hypothesis is that the variation of Sr and Pb isotope composition exhibited by the oceanic alkali basalts is entirely due to isotopic inhomeogeneities in the mantle, and that these inhomogeneities have arisen as a result of local differences in Rb/Sr and U/Pb ratios which have existed in some cases for thousands of millions of years without isotopic exchange or trace element homogenization. This extreme inhomogeneity required by the oceanic islands is in marked contrast to the relatively homogeneous source region inferred for the tholeiites of the world-wide ocean ridge systems. This need for numerous small source regions for alkali basalts, which have maintained hteir separate geochemical identify for periods of order 10^9 years, in some cases in close proximity to the extensive and relatively uniform source region of ridge tholeiites, is the major difficulty with the equilibrium melting model. As Hart has emphasized, it faces us with the problem of understanding how the identities of two distinct mantle sources can be maintained when they are in close proximity and in an area of great mantle mobility.

Disequilibrium Melting

This difficulty, forced on us by the isotope data, is severe enough to warrant investigation of alternative models. One such is disequilibrium partial melting, discussed in the past by Harris et al. [24] and Graham and Ringwood [25]. O'Nions and Pankhurst [14] have recently constructed quantitative models based on disequilibrium melting, and investigated the isotopic and trace element composition of magmas produced by disequilibrium partial melting of reasonable mantle source composition.

The essence of the disequilibrium partial melting hypothesis is that complete isotopic equilibration does not occur between the partial melt and the solid silicate phases in the residue. Since the various silicate phases in the original primitive mantle have different Rb/Sr and different $^{87}Sr/^{86}Sr$ ratios, and since they enter the partial melt at different rates, disequilibrium partial melting affords a mechanism whereby a magma can be generated with a different Sr- or Pb-isotopic composition from its source material. That such a mechanism is not impossible is shown by the work of Leggo and Hutchison [26], who found that lherzolite nodules and their host basalts had different Sr isotopic compositions, whilst Harris [24] cited evidence showing that in one case Sr isotopic exchange between co-existing phases had not occurred during 350 m.y. at temperature of between 600-1000° C. It should therefore seriously be considered whether the observed variation in the isotopic composition of oceanic basalts might in part be due to basaltic magmas forming out of isotopic equilibrium with their source regions due to this process.

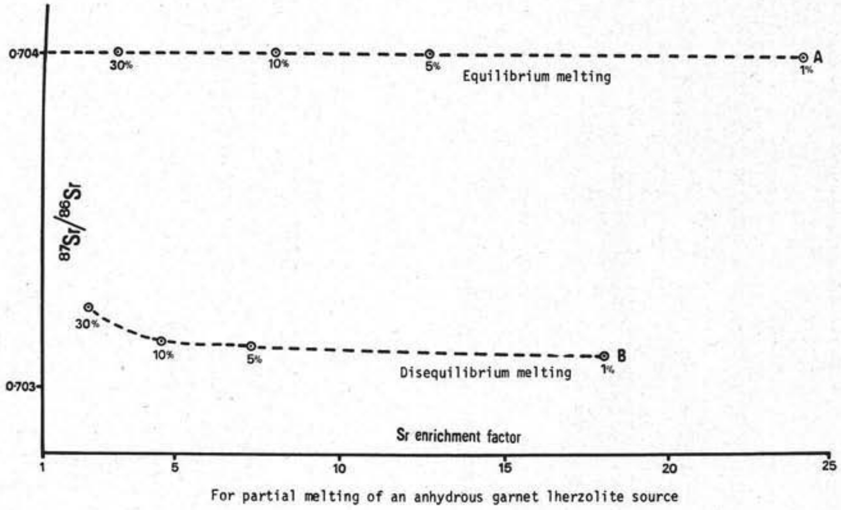


Fig. 11. — Sr-enrichment versus $^{87}Sr/^{86}Sr$ for partial melting of a garnet Iherzolite source. (60 % olivine, 20 % orthopyroxene, 10 % clinopyroxene, 10 % garnet).

Curve A: Equilibrium melting for a source having a bulk $^{87}Sr/^{86}Sr$ ratio of 0.7040. The relative melting rates of olivine, orthopyroxene clinopyroxene and garnet are as 1:5:10:10.

Curve B: Disequilibrium melting. The $^{87}Sr/^{86}Sr$ ratio of olivine, orthopyroxene and clinopyroxene are taken as .7077, .7058 and .7022 respectively. The $^{87}Sr/^{86}Sr$ ratio for the olivine, and all these identical with that for olivine, and all these phases do not completely equilibrate isotopically during partial melting. For other details, see O'Nions and Pankhurst, J. Petrol. *15* (1974), 626.

O'Nions and Pankhurst [14] have made disequilibrium model calculations for partial melting of an anhydrous garnet lherzolite source. Fig. 11 shows the mutual dependence of the $^{87}Sr/^{86}Sr$ ratio of the melt and its Sr enrichment factor; it is clear that, compared with the equilibrium case, a given degree of partial melting produces lower enrichment factors and lower $^{87}Sr/^{86}Sr$ ratios. The most important finding of the model calculations is that disequilibrium melting of a garnet lherzolite source could indeed produce liquids with Sr contents similar to those in alkali basalts (low degrees of partial melting) and abyssal tholeiites (higher degrees of partial melting) but in each case with a *lower* ^{87}Sr/^{86}Sr ratio than the bulk source. This idea was suggested qualitatively by Harris et al. in 1972 [24], but is now substantiated by quantitative calculations.

Quantitative calculations by O'Nions and Pankhurst for radiogenic decay over 20 m.y. of a model source region containing a minor hydrous phase (phlogopite or amphibole) have also been made; the significant difference here is that the ^{87}Sr ^{86}Sr of the liquid is *higher* than the bulk source, largely because the melting of phlogopite (or amphibole) dominates the process.

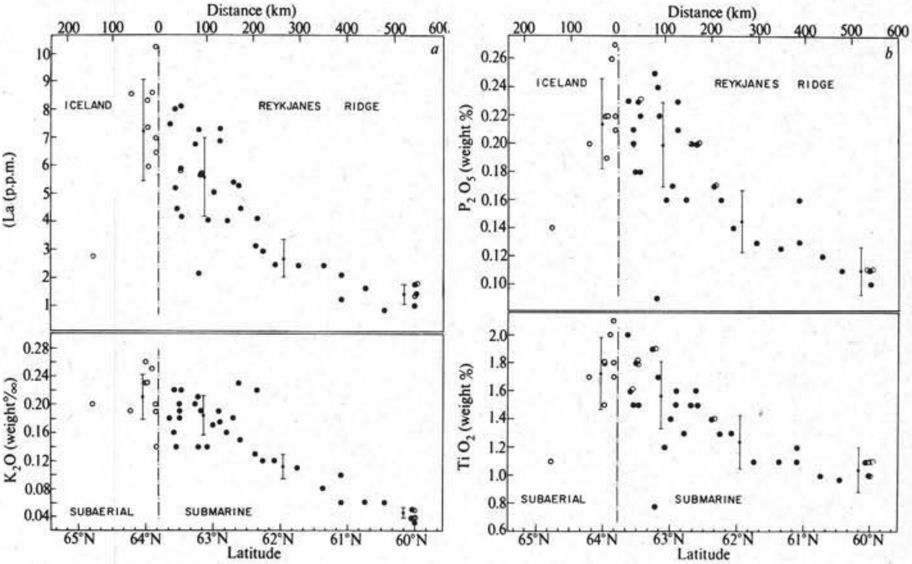


Fig. 12. — La, K$_2$O, P$_2$O$_5$ and TiO$_2$ concentration variations in tholeiitic basalts taken from along the Reykjanes Ridge. The data shows a regular and progressive depletion for these elements from Iceland (where there is a large scatter) down to 61°N, and then levels off until the limit of the survey at 60°N. [From 'Nature *242* (1973, 565].

The general conclusion is that there seems to be adequate scope for disequilibrium partial melting to produce liquids with the full range of ^{87}Sr/^{86}Sr ratios and Sr contents found in oceanic basalts with relatively short periods of closed system behaviour, though not from a single mantle source which is geochemically and mineralogically uniform.

Mantle Plumes

Before summarising the present overall status of isotopic studies in oceanic basalts I must briefly consider the deep mantle plume hypothesis introduced by Morgan [27] and used by Schilling [10], Hart [29] and colleagues to relate island

and ridge volcanics. Morgan introduced the idea of the deep convection mantle plume, tapping regions of the lower mantle and bringing heat and relatively primordial material up to the asthenosphere, largely to provide the motive force for moving plates apart. His model carried with it the assumption that mid-ocean rises are passive, and that when plates are moved apart asthenosphere rises along the line of fracture (the mid-ocean ridge) to fill the void and create the ridge tholeiites. In contrast, under an island like Iceland a plume is assumed to be active, transporting relatively primordial material adiabatically from deep in the mantle upto asthenosphere depths; during the rise partial melting begins, due to decompression melting, and the Iceland lava is essentially all derived from the mantle plume material.

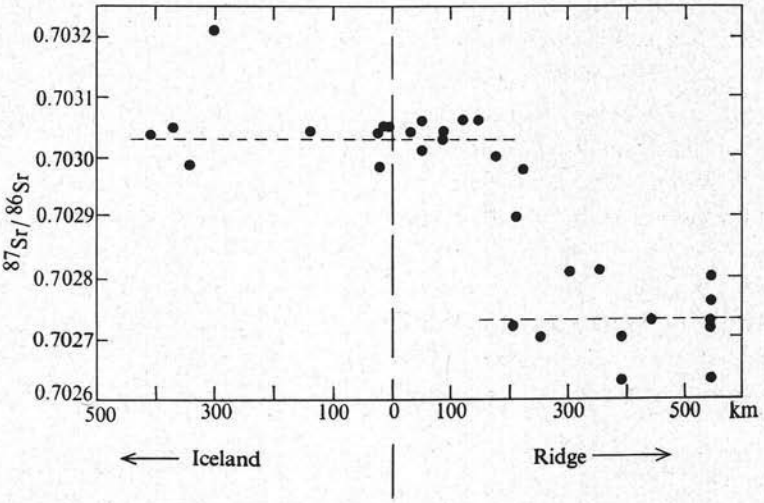


Fig. 13. — Strontium isotope data for the same Reykjanes Ridge tholeiitic basalts as those of Fig. 12. [From Nature Phys. Sci. *246* (1973), 104].

Schilling [10] in 1973 presented data on rare earth and other incompatible element concentrations in samples from the Reykjanes Ridge and its northward extension into Iceland, whilst Hart et al. later presented Sr-isotope data on the same samples. Fig. 12 shows Schilling's trace element data. It is clear that La, K, P and Ti concentrations decrease progressively from Iceland down to 61° N, and then level off to about ridge tholeiite values. This contrasts with but minor variations in major element chemistry, with most of the ridge being quartz tholeiites and the Iceland samples being dominantly olivine tholeiites. Fig. 13 shows the strontium isotope data for the same samples [28]; there are clearly two general groupings of values, with an irregular transition between them. Sixteen samples from Iceland and for 200 km south along the ridge, spanning a distance of 600 km, are identical within 0.01 % at a value of 0.70304. Beyond 250 km south on the ridge the values are consistently lower than on Iceland, averaging 0.7027. This general pattern is confirmed by work at Oxford by O'Nions and Pankhurst [14].

Hart and Schilling have interpreted this data as consistent with the derivation of Iceland tholeiites and Reykjanes Ridge tholeiites from two distinct sources, which mix in the intermediate zone from 200-250 km along the ridge, and as being consistent with, though not compelling evidence for, the plume hypothesis. The remarkable uniformity of Sr isotope ratios in Iceland basalts and those from the first 200 km of the Ridge indicates that these values are primary and represent a particular mantle source. According to the plume hypothesis, its upward flow rate is in excess of that required to create new lithosphere below Iceland, which is also spreading. The overflow of plume material is diverted into the low velocity layer, where it is probably channeled preferentially along the hot, mass deficient and structurally weak Reykjanes Ridge Axis. The plume material will mix with Ridge material proper derived from the asthenosphere; at Iceland plume material dominates, whilst 250 km south along the Ridge asthenosphere dominates.

Recent evidence from Oxford has, however, destroyed the geochemical evidence in favour of the plume hypothesis for Iceland. In the first place Kolbeinsey, only just north of Iceland, has a low $^{87}Sr/^{86}Sr$ ratio, as low as the lowest values obtained for the southern-most end of the Reykjanes Ridge, and so there can be no progressive decrease in $^{87}Sr/^{86}Sr$ along the Mid-Atlantic Ridge north of Iceland comparable to that observed by Hart et al. for the Reykjanes Ridge. Secondly, recent work by O'Nions and Pankhurst on shield volcanoes in the Neovolcanic Zones of Iceland has shown that the whole range in Sr-isotope compositions and REE patterns found along the Reykjanes Ridge can be found within the Neovolcanic Zones of Iceland alone.

What conclusions remain?

1) The $^{87}Sr/^{86}Sr$ ratios of basalts erupted in the Atlantic and Pacific regions show variations which are gross relative to the analytical precision.
2) In general the constancy of this ratio throughout a range of rock types on any one island rules out any significant contamination from older volcanic or crustal rocks — $^{87}Sr/^{86}Sr$ ratios are generally primary characteristics.
3) There is a separation into:-
   (*a*) Ocean ridge tholeiites, with uniformly low $^{87}Sr/^{86}Sr$ ratios and incompatible element contents.
   (*b*) Ocean island tholeiites, which are slightly higher in both.
   (c) Ocean island alkali basalts which have the highest $^{87}Sr/^{86}Sr$ ratios and incompatible element contents.
4) The world-wide, rather uniform, source of ridge tholeiites is relatively depleted in incompatible elements, in Rb relative to Sr, and in U relative to Pb, but the time or times of depletion cannot be determined from Rb-Sr systematics nor

from the presently available Pb isotope data, because of difficulties in solving multistage histories.

5) No definite geochemical constraints can yet be placed on the relative locations of « depleted » and « undepleted » source regions within the mantle. For instance, the rather less depleted character of island tholeiites can be explained by vertical differentiation in which *either* the upper part of the asthenosphere *or* else the deep mantle has higher incompatible element contents and ^{87}Sr/^{86}Sr ratios than those parts of the asthenosphere which are believed to supply mid-ocean ridge magmatism.

6) The wide isotopic variations of island alkali basalts must come from isotopically heterogeneous sources, but the scale of these heterogeneities is not determined and depends on whether equilibrium or disequilibrium melting was involved. *Complete isotopic equilibrium between the mineral phases of the source region* implies that the heterogeneities exist in domains of lateral extent of order tens or hundreds of kilometers — if the Rb/Sr of partial melts is equal to or greater than that of the bulk source, these domains must have remained isolated for periods up to at least 1000 my. This seems improbable in the highly mobile mantle regions involved.

   Disequilibrium partial melting of probable mantle assemblages suggests, on the other hand, that the observed variations of ^{87}Sr ^{86}Sr and trace element contents could be obtained in basaltic melts after quite short periods of closed system behaviour of the constituent minerals.

7) There is at present insufficient geochemical evidence to evaluate the feasibility of the disequilibrium melting hypothesis; in particular diffusion experiments have not been made to test whether interphase isotopic disequilibria can persist under the appropriate partial melting conditions. Such experiments need to be made.

8) There is a clear need for high precision Sr isotope, Pb isotope, and REE data on dredge basalts from as many ridges as possible and on nodule suites from oceanic rocks, since such data is still rather scarce. It is only with more abyssal basalt data that we shall be able to identify trends with certainty, and data on nodule suites may be expected to cast light on the feasibility of disequilibrium melting as a mechanism for ocean basalt production.

I am afraid that it is on that note that I must end. I think that we have to admit that after about ten years of work in this field we have as yet no final answers about the genesis of oceanic basalts and their relation to the mantle. On the other hand both trace element and isotopic data have been able to restrict the number of possible alternatives, and there is the hope that further isotopic work will provide the final key to this problem. The present phase of isotopic and trace element studies on oceanic rocks is undoubtedly a very exciting one, which provides much stimulus for many laboratories to make a concentrated effort to solve the outstanding problems.

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