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The Magnetic and Opaque Petrological Response of Basalts to Regional Hydrothermal Alteration

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Summary

The alteration state of basalt lavas is shown to be largely the result of the combined action of two alteration processes, deuteric oxidation and regional hydrothermal alteration. The former process is well described in the literature while the latter is shown here to be another *widespread* source of lava alteration. The environment of regional hydrothermal alteration is that of the zeolite metamorphic facies, with temperatures of up to 300 °C, and abundant groundwater being the major elements. Independent and rapid spatial variation of the two alteration processes results in the wide range of observed lava properties. Microscopically undetectable titanomagnetite alteration, indicated by steady rise in strong field Curie point, is the first response of a non-deuterically oxidized basalt to regional hydrothermal alteration. With more extreme alteration, titanohematite etc. is seen to pseudomorph titanomagnetite grains. Ilmenite also shows very distinctive mineralogical changes. Initially, high deuteric oxidation specimens appear to be immune magnetically to moderate alteration but these too eventually succumb when conditions are extreme enough for the formation of prehnite in the rock.

The geophysical implications of this widespread post eruption lava alteration process is discussed. We conclude that it is probably possible to obtain information on the original TRM of basalts at least in many cases but that the implications for potassium–argon dating need serious investigation.

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1. Introduction

This paper describes the continuation of investigations by the Liverpool University geophysics group into relationships between the magnetic and petrological properties of basaltic rocks. In this case the investigation was stimulated by the visit of one of the authors (H.C.P.) to Liverpool with an ensuing examination, and discussion of, the complex but fascinating Proterozoic Keweenawan volcanics of the Lake Superior region. We realized that, while the Keweenawan basalts showed many opaque mineral features familiar from Tertiary basalts, such as ilmenite exsolution lamellae in titanomagnetite, they also showed a wide range of new phenomena. These phenomena were clearly later in age than the deuteric oxidation features and we guessed at their origin being in the post-eruptive burial history of the lavas. From this beginning we have been able to collect together petrological (and magnetic) results from a large number of lava sequences and, using depth of burial information obtained from Dr G. P. L. Walker of Imperial College, and other sources, to build up a picture of the progressive magnetic and opaque mineralogical alteration of basalt lavas as a result of post eruption burial by younger flows.

The general result of this study is that the magnetic and opaque petrological properties of any basalt specimen are the result of the combined action of two alteration processes, deuteric oxidation, taking place during initial cooling, and regional hydrothermal alteration, taking place after burial by younger flows. The fact that these two processes vary in intensity independently and rapidly in lava flows at least largely explains the wide range of magnetic properties found for basalts.

The paper is organized to give the descriptive material first, followed by a discussion of the mineralogical and palaeomagnetic implications of the newly described alteration process. As considerable whole-rock chemical changes also take place in response to regional hydrothermal alteration we also consider the implications of this alteration process for potassium–argon dating.

Anticipating the conclusions, it seems likely that information on the original TRM of lavas often can be obtained, following suitable remanence cleaning, notwithstanding the widespread and considerable nature of the alterations to the magnetic minerals. In retrospect, it is hard to believe that palaeomagnetism would have become such a widely studied and respected source of geophysical information had there been previous knowledge of the extent and degree of post-eruptive alteration of basalt lavas.

No such confident conclusion can be made for the influence of regional hydrothermal alteration on the potassium and argon distributions in basalts; this is a field where more experimental work is urgently needed.

2. Zeolites and other secondary minerals as indicators of hydrothermal alteration in basalt lava piles

2.1 *Secondary minerals in lavas*

Mineralogists have long recognized the existence of many apparently late crystallizing minerals in basalt lavas. Of these, the zeolites, here taken as including analcite, form a major group, but various forms of silica, calcite, apophyllite, celadonite, chlorophaeite, prehnite, pumpellyite, epidote and other minerals are also known to occur widely.

The zeolite minerals most characteristically occur in positions in lavas that suggest a late period of formation. Typical localities are interstitially with respect to the primary feldspars and pyroxenes, and in the various cavities (vesicles, amygdules) that mark the positions of gas accumulation during the original lava cooling period. Zeolites are often white or pale coloured and stand out prominently against the typically dark lava. In many cases well-developed crystals are found in partly filled

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vesicles, and these occurrences facilitate identification among the many superficially similar members of the group. Chemically the zeolites are hydrated calcium aluminium silicates, sometimes with sodium and potassium. It is likely that in many cases the source of the zeolite and other secondary minerals forming material is the lava flow itself (Jolly & Smith 1970), their formation marking the partial rearrangement of the chemical constituents of the lava to phases stable in excess water at relatively near surface P–T conditions. Plagioclase feldspar and interstitial glass are obvious sources of zeolite building constituents, and other primary minerals may also contribute. Formation of zeolites almost certainly takes place in an environment where the rock is saturated by warmed meteoric water. Heat can be supplied as a result of burial, from exothermic reactions, by dyke or sill intrusion or by hot magmatic residual fluids from large near surface intrusions.

Of the many non-zeolite secondary minerals found in lavas, only prehnite and epidote are important from the viewpoint of this paper. These two minerals are also largely hydrated calcium alumo-silicates, but differ from the zeolites in containing iron. They tend to be green or yellow in colour.

Two chemical features of lava secondary minerals may have geophysical significance. While zeolites are basically hydrated calcium alumino-silicates, a number of them (for example, stilbite, mordenite (ptilolite) and phillipsite) characteristically contain sodium or potassium. The source of the potassium (and sodium) is probably in the breakdown of pyrogenic minerals or glass. Its relocation in secondary minerals, which can be formed millions of years after eruption, may mean the potassium–argon dates of zeolite bearing basalts refer to the date of zeolite formation rather than the time or eruption of the lava.

Again, the presence of iron in prehnite and epidote probably requires a source in the ferromagnesian or opaque minerals of the lava. If iron is removed from the opaque minerals to form some of these secondary minerals, then long post-eruption changes in the magnetic properties of the opaque iron titanium oxides may be expected to have occurred.

2.2 *The distribution of basalt secondary minerals*

Regional studies of the distribution of secondary minerals in volcanic areas now cover all or parts of Eastern Iceland (Walker 1960b), Northern Ireland (Walker 1960a), Mull, Scotland (Walker 1970) and the Keweenawan volcanics (Stoiber & Davidson 1959), while the distribution in sedimentary piles such as in the Taringatura syncline, Southland, N.Z. (Coombs *et al.* 1959), is also known. From the studies in volcanic areas a picture has emerged of the existence of secondary mineral zones, each characterized by a particular facies of secondary minerals. These zones can be either regional in extent, where they appear to be related to the original surface of the lava pile, or in annular arrangement about volcanic centres. A composite sequence of regional secondary mineral zones has been built up (Fig. 1) in which each zone is described by the name of a single secondary mineral. In fact many secondary minerals occur in each zone and the nominal mineral is only diagnostic in that it first appears at the top of the zone and is then joined by the nominal mineral of the next lower zone at the base of the zone. It should be noted that the letter-zones (A to I) of Fig. 1 do not correspond to the zeolite-nominal zones. This is unfortunate, but arises from a reasonable attempt to divide one of the basic sections, the Eastern Iceland lava pile, into similar depth of burial intervals, using the top of the analcite zone as datum. It must be stressed that use of this zonal scheme is not intended to imply that temperature gradients near to the Earth's surface are uniform. However, use of the scheme does allow a first attempt at relating basalt secondary mineral development, regional reheating resulting from burial, and associated magnetic and mineralogical changes to basaltic opaque minerals.

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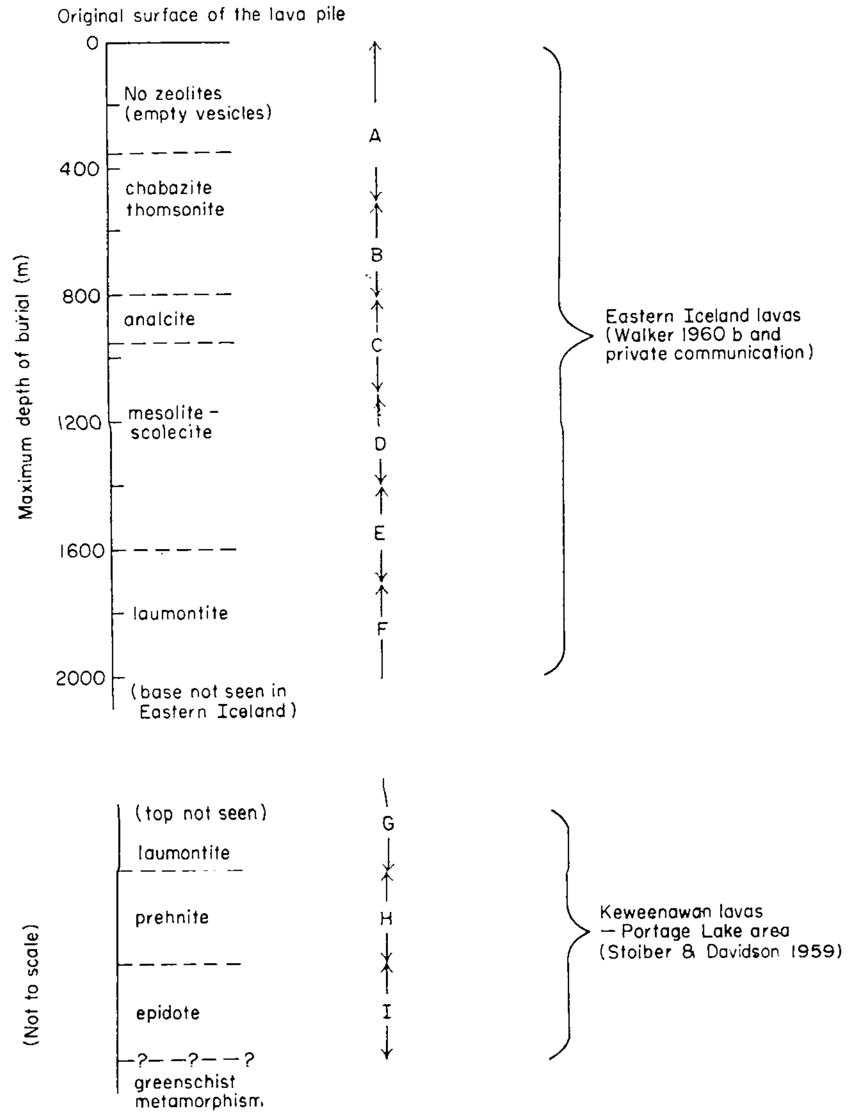

FIG. 1. Regional secondary mineral zones. The standard succession based on the Eastern Iceland and Keweenawan lava piles.

2.3 *Chemical and physical conditions during secondary mineral formation*

For consideration of both magnetic changes and argon loss possibilities we need to know the chemical and physical history of a rock. Here we are concerned with the chemical and physical history of basalts as indicated by their secondary mineral contents. The chemical changes observed represent the partial replacement of quenched anhydrous high temperature phases by phases stable in the presence of excess water at no more than a few hundred degrees centigrade and probably less than 4 kb pressure.

Elucidation of the pressure–temperature history of basalts can be attempted from both field and laboratory observations. The vertical zonation of zeolite distribution in lava piles is consistent with the formation of these secondary minerals being both or either temperature or pressure dependent. The annular zonation of prehnite

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and epidote around intrusive centres may imply that the temperature at which they first form is little influenced by pressure variation over the pressure range in question.

Not all of the important minerals have been studied in the laboratory but stability information does exist for a sufficient number of secondary minerals to allow an attempt to be made to temperature-calibrate the field zone sequence. It is clear from the literature that the experimental definition of stability fields for these low temperature secondary minerals is a difficult task. This is at least partly the result of the necessity to work over temperature ranges where achievement of equilibrium may take very long periods. In the several important cases there is no certain definition of the stability field and we can only summarize the results and opinions of the workers in the field.

Chabazite and Phillipsite—the first appearance of zeolites in lava successions. There seems to be little doubt that these phases can form at temperatures close to or a little above the ambient. Authigenic phillipsite has been reported as occurring in calcareous deep sea sediments by Young (1939) while Daubree (1879) has described both chabazite and phillipsite in the wall work of the Roman Baths at Plombieres and elsewhere. The water temperatures in these Baths range from 49 °C to 70 °C.

The lower limit of analcite stability. This important parameter does not seem to have been examined in any specific investigation. The only evidence found in the literature refers to the formation of analcite in rhyolitic and dacitic lavas in a geyser basin in Yellowstone Park (Fenner 1936). Here analcite is forming at between 125 °C and 155 °C in an alkaline environment from 26 to 60 m below the surface. While the conditions of formation of analcite at Yellowstone are probably rather different from conditions in basalts we will use a mean temperature of 140 °C for a provisional location of the reference point for this mineral.

Laumontite and the upper limit of analcite stability. The stability fields of these two minerals are considered together following Winkler's 1967 definition of the laumontite–prehnite–quartz facies. According to Winkler the reactions defining the low temperature limit of this facies are:

$$\text{Heulandite} \rightarrow \text{Laumontite} + \text{Quartz} + \text{Water} \quad (1)$$

and

$$\text{Analcite} + \text{Quartz} \rightarrow \text{Albite}. \quad (2)$$

Early attempts at determining the equilibrium temperature for reaction (1) at pressures within the range of interest were described by Coombs *et al.* (1959) and Winkler (1965). Coombs *et al.* suggested that the equilibrium temperature was lower than 280 °C at 2 kb while Winkler proposed a small variation about 280 °C over the pressure range from 0·5 to 4 kb. More recent work suggests rather lower equilibrium temperatures. Senderov (1965) describes laumontite formation from alkaline fluids in dacitic tuffs in an active hydrothermal system in Kamchatka at temperatures between 100 °C and 180 °C. Hietanen (1967) suggests temperatures of between 160 °C and 170 °C at less than 1 kb, and Nitsch (in Winkler 1967) states that heulandite is stable at up to 200 °C at 7 kb, with equilibrium temperature decreasing with reduced pressure.

Reaction (2) has been studied by Campbell & Fyfe (1965) and Althaus (rep. in Winkler 1967). Campbell & Fyfe suggest that the upper limit stability field of analcite is highly pressure dependent. Over the pressure range 0–2 kb the equilibrium temperature for reaction (2) ranges between 170 °C and 190 °C while by 4 kb it has dropped to ambient temperatures. In contrast, Althaus finds experimentally that the equilibrium in (2) is at close to 220 °C over a pressure range of 4 kb to 7 kb.

Finally Turner (1968) indicates that the zeolite facies should be replaced by the prehnite–pumpellyite assemblage at about 170 °C for pressures of up to 2 kb.

Clearly, from the different results listed above, there is no final agreement of the temperature of this important boundary in the relevant pressure range. For the

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purpose of this paper we propose to take a representative temperature of 170 °C with the proviso that this figure may well have to be modified when the position of the boundary is finally agreed.

Prehnite. The lower limit of the stability field of prehnite has been the subject of several investigations with a considerable degree of agreement between different results. Fyfe (1955) produced prehnite from a glass of prehnite composition at temperatures down to a little below 300 °C. It seems from this investigation and later investigations by Coombs *et al.* (1959) that prehnite forms most readily at pressures of above 2 kb, a fact which should be kept in mind in the interpretation of secondary mineral zones. Later investigations by Hietanen (1967), Nitsch (in Winkler 1967) and Strens (1968) all confirm a lower limit of close to 300 °C, with a 2 kb pressure quoted for Stren's result. The only major divergence from the 300 °C lower limit for the prehnite stability field is found in Turner (1968) where our perhaps unjustified extrapolation to low pressures in Turner's figure yields a limit of from 180 °C to 240 °C.

Epidote. As in the case of prehnite there appears to be a degree of unanimity concerning the lower limit of the epidote stability field. Stringham (1952) suggested a figure of 300 °C while Winkler & Nitsch (1963), Strens (1964) and Müffler & White (1969) all find similar temperatures, with pressures ranging from near ambient to at least several kilobars. The only major divergence from the widely-determined figure of 300 °C is the record in Strens (1965) of the formation of epidote at between 150 °C and 250 °C at low pressures in thermal areas.

Comparison of the results of the stability field determinations for prehnite and epidote suggests that a conflict exists between the laboratory results and the findings of the field mapping of secondary mineral zones. In the field a prehnite zone can at least in some cases be distinguished from a higher temperature epidote zone (Walker 1970) whereas laboratory results suggests that the minerals have close lower temperature limits to their stability fields. For the purpose of this paper we propose to resolve this apparent conflict by assuming the two stability fields are separated by a few tens of degrees centigrade and we take the lower limit of the prehnite field at 300 °C. As in the case of other secondary mineral stability fields this decision will probably need modification in the future.

Table 1

Lower temperature limits of stability fields over relevant pressure ranges for basalt secondary minerals defining levels in the zonal scheme

Secondary mineral(s)	Temperature (°C)
Chabazite-phillipsite	Just above ambient
Analcite	140
Laumontite	170
Prehnite	300

Table 2 shows the approximate locations within the secondary mineral sequence corresponding to reheating temperatures of 100 °C, 200 °C and 300 °C.

Table 2

Location in the secondary mineral sequence of reference reheating temperatures

Maximum reheating temperature (°C)	Zonal position
Ambient	Top of the zeolite free zone (zone A)
100	Near the base of the chabazite–thomsonite zone (zone B)
200	Just below the top of the laumontite zone (near the E/F boundary)
300	At the laumontite–prehnite (G/H) boundary

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The need now arises for a means of checking the reasonableness of actual crustal temperature gradients based on the reference points. For example, a temperature gradient of about 115 °C/km is implied for Eastern Iceland lava pile at the time of zeolitization, this being about four times larger than the much quoted average gradient of 30 °C/km for a geologically wide range of areas (Fig. 2). A source of independent comparison data is the direct measurements of temperature gradients in non-thermal areas in Iceland by Bodvarsson & Palmason (1961). Five geographically separate measurements give a range of values of from 33 °C/km to 165 °C/km, with a mean of 82 °C/km. These values bracket our estimated gradient, and encourage belief in its reasonableness as an estimated temperature gradient for Eastern Iceland during its period of volcanic activity in the Neogene and Quaternary. Fig. 2 also shows the estimated temperature gradient for the thick Taringatura sedimentary pile in New Zealand. This gradient is estimated to be about 24 °C km, indicating that a range of fossil regional temperature gradients and thus secondary mineral zone thicknesses of at least a factor of five may be expected.

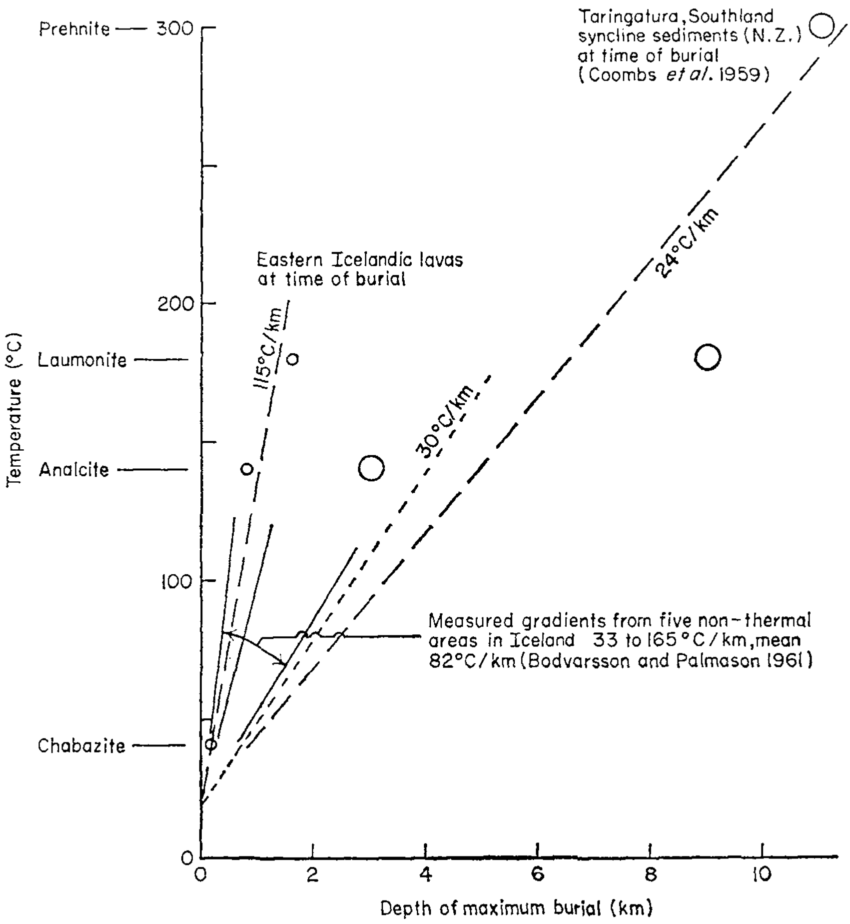

FIG. 2. Thermal gradients in volcanic and sedimentary areas.

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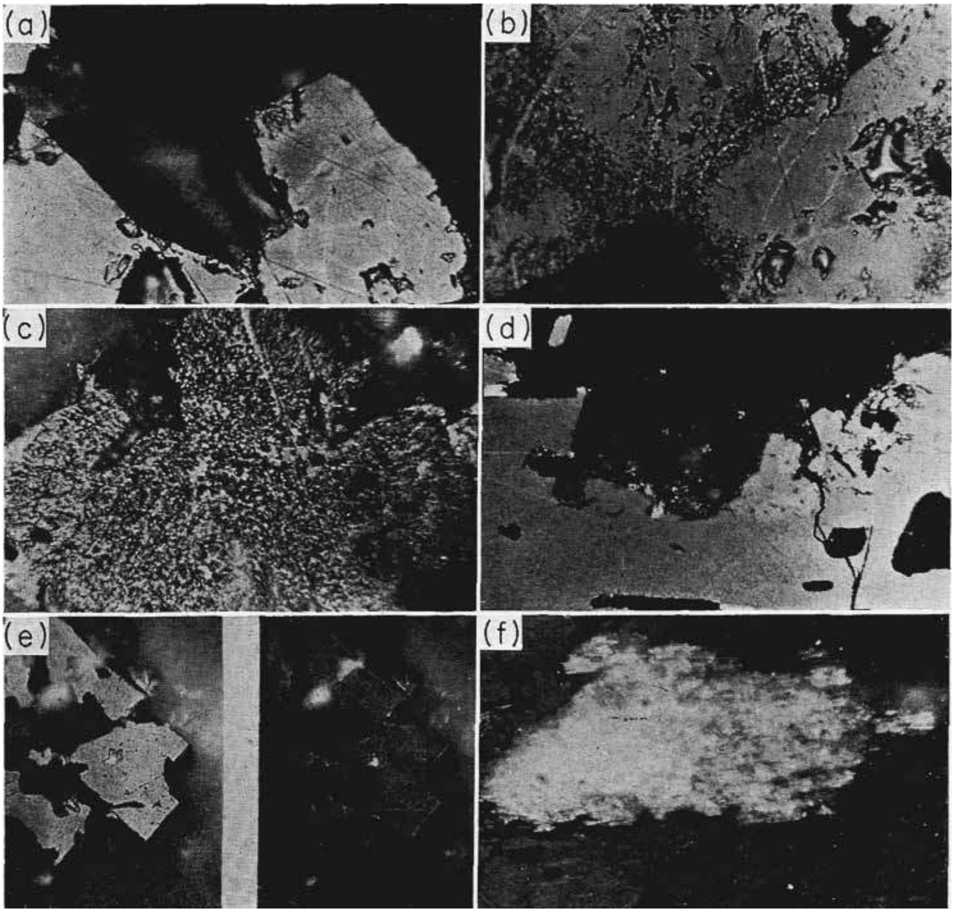

FIG. 3. Progressive regional hydrothermal alteration of deuteric oxidation Class 1 titanomagnetite. (a) Homogeneous titanomagnetite with occasional maghemite lined cracks (Skye lava, between zones B and D). (b) Development of peripheral and crack margin granulation (Skye lava, between zones B and D). (c) Overall granulation with formation of titanohematite from small remaining islands of titanomagnetite (Keweenawan MP19, zone I). (d) Formation of titanohematite flecks in titanomagnetite (Keweenawan D1-6, zone I). (e) Titanohematite aggregate pseudomorph after titanomagnetite; left: plane polarized light; right: polars 25° from crossed position—the different crystallographic orientation of the components of the titanohematite aggregate is seen (Keweenawan MP 45, zone I). (f) Golden yellow sphene replacement of magnetite (Caledonian granite specimen 939).

* Scale: In order to illustrate different features the opaque grains shown in Figs 3-8 are not *all* to the same scale. However, *all* the grains are within a size range typical for many basalt rocks (i.e. from 30 to 100 μ linear dimension).

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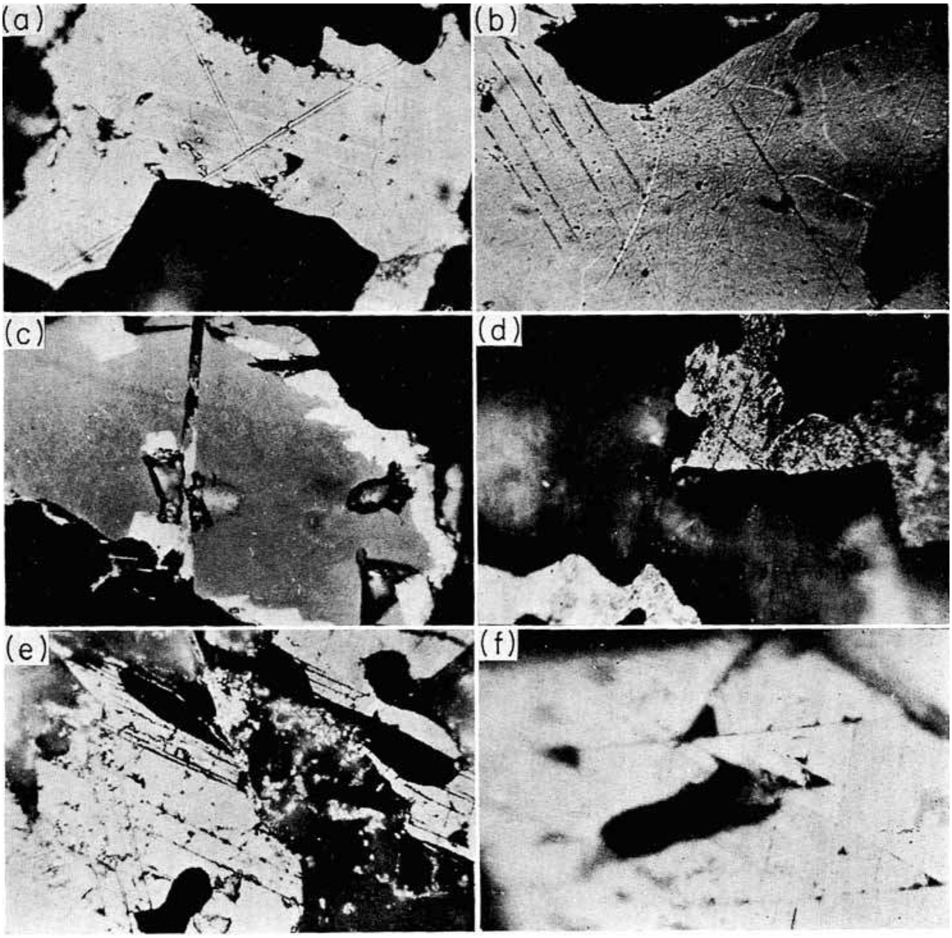

FIG. 4. Progressive regional hydrothermal alteration of deuteric oxidation Class 2 titanomagnetite. (a) Deuteric oxidation Class 2 titanomagnetite without visible evidence of regional hydrothermal alteration. Medium grey-titanomagnetite, off white-ilmenite lamellae (Skye lava, between zones B and D). (b) Class 2 titanomagnetite with the occasional thin ilmenite lamellae now pseudomorphed by black iron bearing sphene (Keweenawan specimen MP73, zone I). (c) Class 2 titanomagnetite with peripheral hematization (white) and a partially decomposed ilmenite lamellae. Note replacement of the lamellae is both by black sphene (upper part) and the 'woven' arrangement of light and medium grey phases (see text) in the lower part (Caledonian granite 941). (d) Highly granulated Class 2 titanomagnetite with black sphene replacement of the occasional original ilmenite lamellae (Keweenawan D1–5, zone I). (e) Titanohematite aggregate pseudomorph after titanomagnetite with black sphene pseudomorphs after the original thin ilmenite lamellae. (f) Note the control on the elements of the titanohematite aggregate by the lamellae (Nuneaton volcanics, NA3-1).

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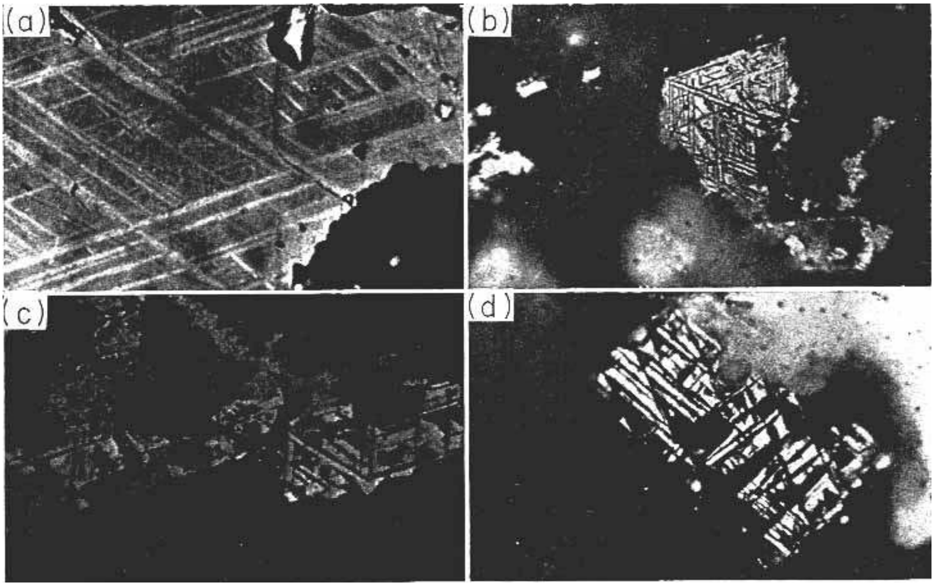

FIG. 5. Progressive regional hydrothermal alteration of deuteric oxidation Class 3 titanomagnetite. (a) Class 3 titanomagnetite without visible evidence of regional hydrothermal alteration. Medium grey, magnetite, light grey; lamellae of exsolved ilmenite (Skye lava, between zones B and D). (b) Black sphene pseudomorphing of original ilmenite lamellae in a Class 3 magnetite (Mull lava A03C200–3, zone I). (c) Regional hydrothermal alteration of a compound titanomagnetite (lower) and ilmenite (upper) grain. The titanomagnetite (dark grey) is partially replaced by titanohematite (off white) while the original ilmenite lamellae are now entirely pseudomorphed in black sphene. The ilmenite component of the compound grain is now entirely pseudomorphed in a fine ' woven ' arrangement of a light and a medium grey phase (Keweenawan MP39, zone I). (d) Class 3 titanomagnetite with the magnetite now entirely replaced by titanohematite (presumably close to pure hematite) and the ilmenite lamellae replaced by black sphene (Keweenawan D12–5, zone I).

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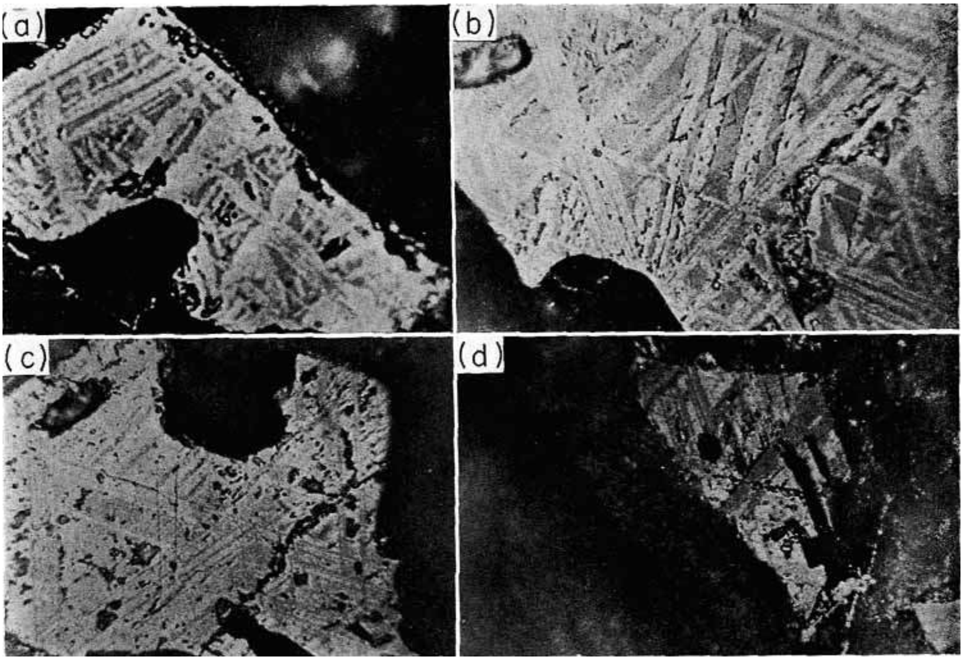

FIG. 6. Progressive regional hydrothermal alteration of deuteric oxidation Class 4 titanomagnetite. (a) Class 4 titanomagnetite without visible evidence of regional hydrothermal alteration. Medium grey-remaining magnetite; off white: a fine aggregate of titanohematite and rutile replacing the original ilmenite of exsolution lamellae. Note the swollen pod like form of many lamellae pseudomorphs (Skye lava, between zones B and D). (b) Typical Class 4 grain persisting unaltered in lavas that have experienced zone I alteration conditions (Keweenawan MP18, zone I). (c) Regional hydrothermal alteration of Class 4 magnetite. Any remaining magnetite has been hematized (off white and light grey areas) while the remains of titanohematite/rutile pseudomorphs after ilmenite lamellae, defining the Class 4 status, are seen as trains of grey dots (Keweenawan MP45, zone I). (d) Regional hydrothermal alteration of Class 4 magnetite. Off white: hematized magnetite, grey: rutile/titanohematite pseudomorphs after original ilmenite lamellae (Keweenawan MP59, zone I).

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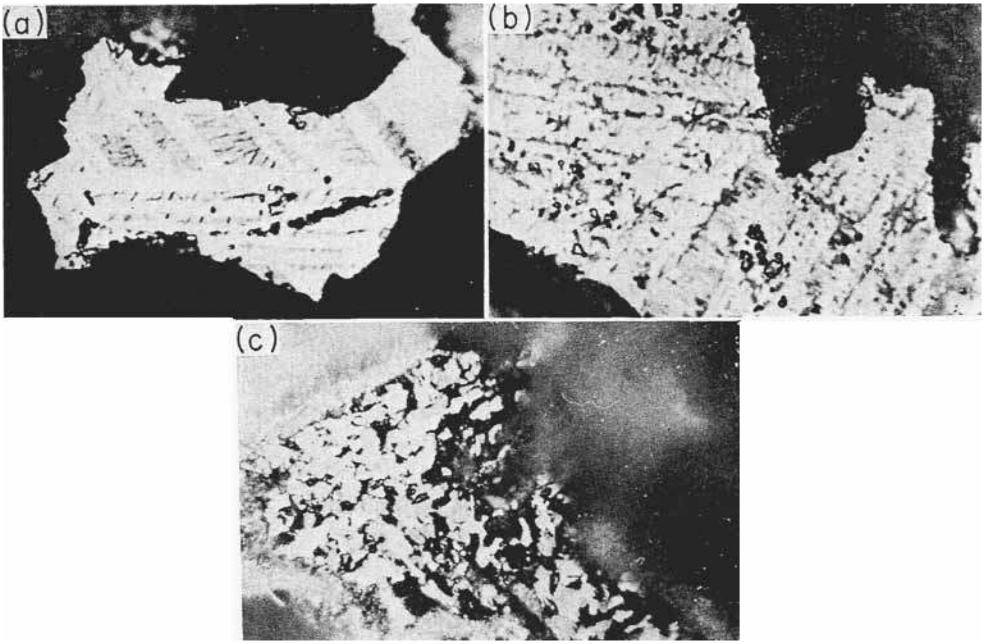

FIG. 7. Class 5 and Class 6 titanomagnetite. (a) Class 5 titanomagnetite without visible evidence of regional hydrothermal alteration. Medium grey; magnetite, black; spinel spicules exsolved from magnetite, off white; pseudomorphs after ilmenite lamellae consisting of fine aggregates of rutile and titanohematite (Skye lava, between zones B and D). (No definite identification of a regionally hydrothermally altered Class 5 grain has yet been made.) (b) Class 6 pseudomorph after titanomagnetite without visible evidence of regional hydrothermal alteration. Off white; titanohematite, medium grey; rutile, dark grey; pseudobrookite. Note how the arrangement of the phases suggests that an octahedrally directed set of ilmenite exsolution lamellae was present at an earlier stage in the oxidation history of the grain (Skye lava, between zones B and D). (c) Probable Class 6 grain from the zone I Keweenawan lavas (MP67), off white; titanohematite, medium grey; probably a pseudobrookite.

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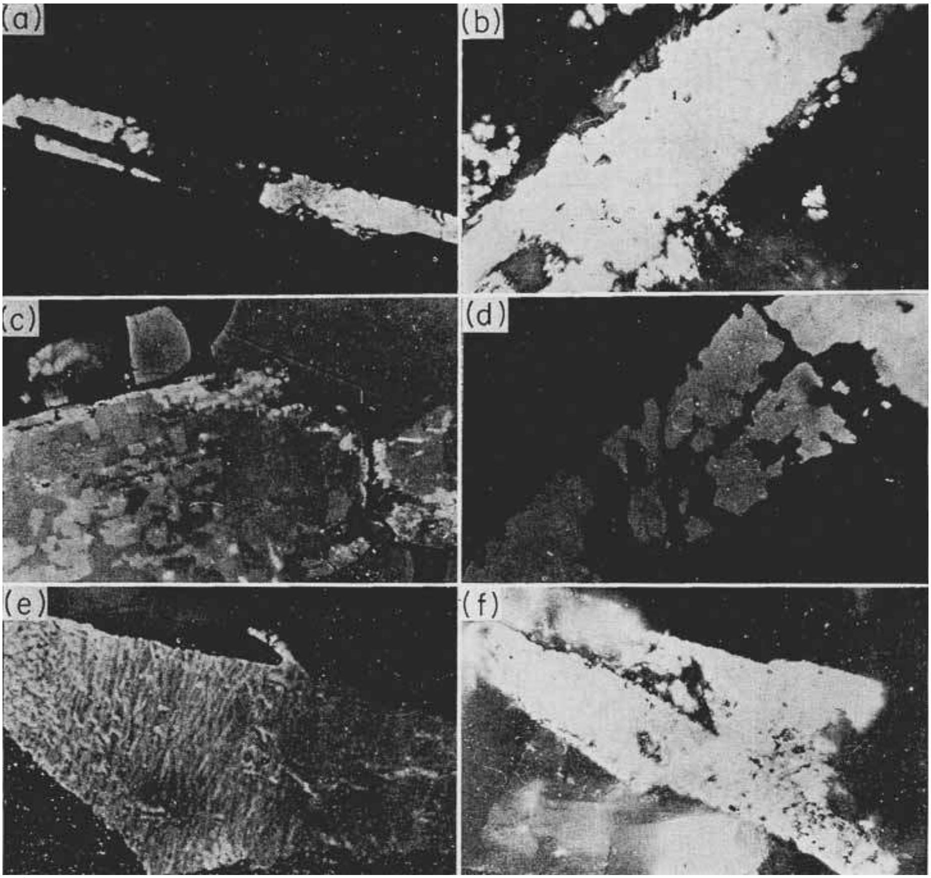

FIG. 8. Progressive regional hydrothermal alteration of non-deuterically oxidised separate ilmenite grains ((a) to (d)). The deuteric oxidation of separate ilmenite is shown in (e) and (f). (a) Non-deuterically oxidized ilmenite without visible evidence of regional hydrothermal alteration. The elongated shape is common (Skye lava, between zones B and D). (b) Regional hydrothermal alteration of a non-deuterically oxidized separate ilmenite grain. Medium grey; ilmenite, mottled patches within grain; woven textural arrangement of high and low titanium replacement products of ilmenite, dark grey to black peripheral alteration phase is iron bearing sphene (Mull lava A03C110–1). (c) Regional hydrothermal alteration of a separate ilmenite grain in a cluster of ilmenite and magnetite grains. The ilmenite grain is on the lower left. Note the interior alteration with the woven texture and the marginal selvage of black sphene adjacent to the magnetite grain on the upper right (Caledonian granite 937). (d) Regional hydrothermal alteration of a separate ilmenite: medium grey; ilmenite, black; peripheral alteration to sphene (Caledonian intrusive 939). (e) Deuteric oxidation of separate ilmenite without visible indication of regional hydrothermal alteration. Medium grey; ilmenite, off-white; ferri-rutile (Skye lava, between zones B and D). (f) Deuteric oxidation of separate ilmenite to a higher degree than seen in (e). Off white; titanohematite, medium grey; rutile, dark grey; pseudobrookite. Although this grain has experienced zone I alteration conditions its mineralogical characteristics are typical of many unburied highly deuterically oxidized separate ilmenites (Keweenawan MP16, zone I).

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3. Changes in opaque mineralogy due to regional hydrothermal alteration

3.1 *Introduction*

In this section mineralogical situations arising from various combinations of deuteric (immediately post eruption) high temperature oxidation and much later regional hydrothermal alteration, are analysed, with the emphasis being placed on the description of newly recognized regional hydrothermal alteration products. Regional hydrothermal alteration produces a number of new iron oxide phases from basaltic titanomagnetite, ilmenite and olivine and concomitant changes in magnetic properties may be expected. However, for clarity, we have decided that this section shall consist largely of a description of the observed mineralogical changes while the description and discussion of magnetic and other changes will form Sections 4 and 5 respectively.

3.2 *Deuteric (high temperature) oxidation of basaltic rocks*

Here we give a resumé of the now rather well known oxidation process occurring in basaltic rocks in the 800–500 °C interval of the initial cooling. This process has been investigated in the field from oxygen fugacity measurements in lava lake drill holes (Sato & Wright 1966), by the methods of experimental petrology (Lindsley 1962, 1965) and so far as the changes produced in the opaque minerals are concerned, in a series of combined ore microscope and magnetic investigations (Wilson & Haggerty 1965, Wilson and Watkins 1967, Grommé, Wright & Peck 1969).

While a wide range of mineralogical changes is produced by this high temperature oxidation process, our concern is only with the changes affecting the original magnetic phase, titanomagnetite, and other minerals where their oxidation products are likely to be ferrimagnetic. The oxidation products of ilmenite and olivine are most interesting from this latter viewpoint. Two features of the deuteric oxidation of basalts are significant here. Firstly, it has been possible using the titanomagnetite of basalts to construct a scale of degree of deuteric oxidation with six steps. States (or classes) of titanomagnetite oxidation illustrating these steps as defined by Ade-Hall *et al.* (1968) are shown in Figs 3–7. Secondly, the degree of deuteric oxidation typically varies considerably within a unit. Variation within one unit may occur with a wide range of characteristic dimensions. Thus, part of a unit may contain a broad zone of high deuteric oxidation, perhaps several metres thick, while within this zone individual opaque grains only a few tens of microns apart may differ by several steps in their degree of deuteric oxidation. The existence of this small scale variation has necessitated the definition of an average titanomagnetite oxidation state (magnetite oxidation number) for a single small specimen (Wilson, Haggerty & Watkins 1968). This number runs continuously from 1·00 (no oxidation) to 6·00 (maximum oxidation).

3.3 *Regional hydrothermal alteration of basaltic rocks*

A most important consideration to bear in mind here is that the regional hydrothermal environment will have acted on a wide range of different types of opaque grains; not only the titanomagnetite and ilmenite of initial crystallization, but all the different steps produced from these minerals by varying degrees of deuteric oxidation. Thus, a basaltic rock that has experienced both deuteric oxidation and regional hydrothermal alteration may be expected to have a most complex opaque mineralogy.

The way in which we have chosen to present this complex picture is taking each deuteric oxidation step in turn, to describe the regional hydrothermal products, where they are known, in order of increasing degree of hydrothermal alteration.

3.3.1 *Titanomagnetite of Class* 1 (*step* 1) (*Fig.* 3). This is the magnetic opaque mineral of non-deuterically oxidized, non-regionally hydrothermal altered (Zone A) basalts. The titanomagnetite is a uniform light brown colour; microprobe measurements show compositions to be clustered about $0 \cdot 3Fe_3O_4 . 0 \cdot 7Fe_2TiO_4$. Alterations

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not specifically related as yet to either of the recognised alteration processes include patchly lightening in colour (while still remaining isotropic) and the formation of linings of an isotropic off-white phase along characteristically curved cracks (Fig. 3(a)). In both cases a type of titanomaghemite may be the alteration product.

The influence of regional hydrothermal alteration is first seen in the appearance of finely divided ferri-rutile granules (Ade-Hall *et al.* 1968) (Fig. 3(b)). Patches of these granules produce a texture described as granulation. In the Eastern Iceland lavas widespread granulation first appears in Zone D, corresponding to a temperature of about 150 °C (Fig. 1), while for a Skye lava studied in detail (Ade-Hall *et al.* 1968) granulation may have commenced at a lower temperature. Granulation, as with other deuteric and regional hydrothermal alteration effects, may vary considerably in its development from grain to grain within one unit. Thus, in the Skye lava just referred to, some Class 1 magnetites were completely ungranulated, others showed partial granulation, with the alteration obviously spreading from grain margins or cracks (Fig. 3(b)), while others were completely granulated as shown in Fig. 3(c). For reasons not known to us the titanomagnetite of some units remains completely ungranulated, even when other indications show that regional hydrothermal alteration in excess of Zone D conditions has taken place. Again, in units where granulation has commenced, increasing regional hydrothermal alteration does not always result in completely granulated grains; it is as if the process of production from titanomagnetite of fine ferri-rutile granules is replaced by other processes at higher temperatures.

Next we see the development of fine white flecks in the titanomagnetite (Fig. 3(d)). These are seen in specimens which have experienced Zone I (>300 °C) conditions. In contrast with the phase of the white alterations described above, the phase responsible for these white flecks is anisotropic and is probably a titanohematite. Increasing alteration of this type results in the agglomeration of the white flecks to form areas of off-white or pale grey pleochroic, anisotropic material. Complete pseudomorphing of original titanomagnetite by a polycrystalline aggregate of this phase is quite frequent after experiencing Zone I conditions (Fig. 3(e)). Microprobe analysis of the grain in Fig. 3(e) shows no compositional variation from area to area within the grain and an overall iron to titanium ratio rather close to that for homogeneous (Class 1) titanomagnetites of Zone A.

Finally, also in specimens that have experienced Zone I regional hydrothermal alteration conditions, complete replacement of titanomagnetite by a bright golden non-opaque phase is occasionally found (Fig. 1(f)). This may be a form of sphene (Haggerty 1968). At first the equivalence of these pseudomorphs with original titanomagnetite was not recognized, but later identification of the relics of octahedral exsolution patterns confirmed this correlation.

3.3.2 *Titanomagnetite of Class* 2 (*Fig.* 4). This deuteric oxidation class represents the onset of visible deuteric oxidation in basaltic titanomagnetite. A small number of rather pure ilmenite lamellae, restricted in orientation to directions parallel to the octahedral planes of the titanomagnetite host, are seen (Fig. 4(a)). Regional hydrothermal alteration produces distinctive mineralogical alteration combinations in both the host titanomagnetite and the ilmenite lamellae.

The titanomagnetite alteration states are those already described for Class 1 grains: granulation (Fig. 4(d), titanohematite flecking and complete replacement by polycrystalline aggregates of titanohematite (Fig. 4(e) and (f)). Fig. 2(c) shows a granitic magnetite, with peripheral hematization which is rarely seen in basalts. This grain (together with grains shown in Figs 3(f), 8(c) and (d)) was chosen to illustrate alteration features which are also typical of basalts, in this case, the nature of the breakdown of an ilmenite lamella.

The regional hydrothermal alteration of the ilmenite lamellae of Class 2 titanomagnetites produces visually striking mineralogical combinations. The most striking

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phase produced has a dark blue-black colour, for which microprobe measurements suggest an iron bearing sphene (sphene–calcium titanum silicate, $CaTiSiO_5$) composition. The calcic nature of this phase supports the necessity for there to have been considerable migration of ions within the rock, as already suggested by the presence of calcic zeolites; since non-hydrothermally altered Class 2 basaltic magnetite is essentially calcium free the calcium of the iron-bearing sphene must be derived from the breakdown of plagioclase felspar, or perhaps, calcic pyroxene. Fig. 4(b), (c), (d), (e) and (f) all show examples of the blue-black sphene replacement of original deuteric oxidation produced ilmenite lamellae. Replacement of ilmenite lamellae by sphene is first seen in Zone G lavas (from Mull). Zone G consists of the lower part of the laumontite zeolite zone for which maximum regional hydrothermal alteration temperatures are probably between 250 °C and 300 °C.

Several other phases also replace ilmenite lamellae. Some of these (? titanohematite, ? ferri-rutile) are seen in the decomposing ilmenite lamellae of Fig. 4(c). In addition, a granulation type of alteration is sometimes seen. Alteration of ilmenite lamellae (and of separate ilmenite) to a combination of possible titanohematite and possible ferri-rutile, described in detail later, is a feature of Zone I alteration (>300 °C)

3.3.3 *Titanomagnetite of Class* 3 (*Fig.* 5). Deuteric oxidation Class 3 titanomagnetite only differs in ilmenite lamellae total area from Class 2 titanomagnetite. In a Class 3 grain lamellae formation approaches the maximum possible, where half or more of the grain now consists of lamellae (Fig. 5(a)). Consequently the remaining magnetite closely approaches pure magnetite, Fe_3O_4, in composition.

Regional hydrothermal alteration of Class 3 magnetite shows many of the features already described for Class 2 and Class 1 magnetite. Thus, partial hematization of magnetite (Fig. 5(c)) and blue-black sphene replacement of original ilmenite lamellae (Fig. 5(b), (c) and (d)) are characteristic of the higher stages of alteration. However, in contrast with Class 1 and 2 magnetite, granulation is rare or completely absent in Class 3 magnetite (see Ade-Hall *et al.* 1968, part I, Fig. 5). This is consistent with the nearly complete removal of titanium from the original titanomagnetite during deuteric ilmenite lamellae formation.

3.3.4 *Titanomagnetite of Class* 4 (*Fig.* 6). Deuteric oxidation Class 4 magnetites are distinguished from Class 3 magnetites by further oxidation causing the breakdown of ilmenite lamellae to fine aggregates of titanohematite and ferri-rutile. This results in a whitening of the lamellae and a volume increase, causing the lamellae frequently to show pod-like form (Fig. 6(a)). The regional hydrothermal alteration of Class 4 magnetite to Zone I conditions produces rather irregular to pod-like relic lamellae in a hematite base, the lamellae apparently still consisting of a combination of finely divided titanohematite and ferrirutile (Fig. 6(c) and (d)). Fig. 6(b) shows a Zone I Class 4 grain with the magnetite still preserved. The grains of Fig. 6(a), (b) and (c) all derive from the Keweenawan Mamainse Point lavas (Zone I), again illustrating the variability of degree of regional hydrothermal alteration in a limited area.

It is convenient at this point to bring to the reader's attention the fact that titanohematite and ferri-rutile apparently can be produced from basaltic iron–titanium oxides in both the deuteric oxidation and the regional hydrothermal alteration environments. At this stage in our knowledge textural considerations must be used to separate chemically similar products of the different environments. It remains to be seen whether microprobe or other analysis will show compositional differences within solid solution series between the phases produced in the different environments.

3.3.5 *Titanomagnetite of Class* 5 (*Fig.* 7). Deuteric oxidation of Class 5 consists of distinctive grains formed of islands of honey-brown magnetite containing black spinel lamellae and separated by broad oxidized lamellae (Fig. 7(a)). At present

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we are not aware of the effect of regional hydrothermal alteration on this type of grain.

3.3.6 *Titanomagnetite of Class* 6 (*Fig.* 7). Deuteric oxidation of titanomagnetite to Class 6 produces a pseudomorph from which magnetite is completely absent. Characteristically, the pseudomorph consists of titanohematite, ferri-rutile and pseudobrookite, or just titanohematite and one of the latter two Ti bearing phases. The distribution of these oxidation products of magnetite and ilmenite are often related to the octahedral pattern of the original ilmenite lamellae (Fig. 7(b)). Regional hydrothermal alteration of basalts may leave Class 6 grains in an apparently little altered form or may produce the type of grain illustrated in Fig. 7(c). This type of grain consists of an aggregate of roughly equidimensional areas of white and grey phases, with probe-determined compositions consistent with titanohematite and ferri-rutile or a pseudobrookite. However, the lack of a recognizable magnetite-derived texture must leave the classification of this type of grain in doubt. Alternatively it could represent an extreme form of the regional hydrothermal alteration of separate ilmenite (see Section 3.3.7).

3.3.7 *Separate ilmenite* (*Fig.* 8). Separate grains of ilmenite, that is, those formed during initial crystallization, show several types of deuteric alteration. For the purpose of classification only two stages are recognized, non-deuterically altered separate ilmenite, and, including several types of alteration together, deuterically oxidized separate ilmenite.

Non-deuterically altered separate ilmenite shows a very clear mineralogical response to regional hydrothermal alteration. The homogenous brown separate ilmenite of Fig. 8(a) shows, after Zone I conditions, partial replacement by several different phases. Characteristically peripherally, suggesting chemical exchange with the rest of the rock, there is irregular alteration to a dark blue-black iron bearing sphene (Fig. 8(b), (c) and (d)). Internally within the same grains patches consisting of roughly equidimensional areas of a white and a light grey phase often begin to replace the ilmenite (Fig. 8(b) and (c)). Electron microprobe measurements indicate that the Fe : Ti ratio is about twice as high in the white phase than it is in the light grey phase. We propose to describe this arrangement of phases as ‘ woven ’ texture on the grounds of its resemblance to a woven material.

Fig. 8(e) shows a deuterically oxidized separate ilmenite grain, with typically curved possibly ferri-rutile exsolution bodies oriented roughly across the short dimension of the grain. Fig. 8(f) shows a deuterically oxidized separate ilmenite later subjected to Zone I regional hydrothermal alteration conditions; although this grain is different from that shown in Fig. 8(e), the new phases seen are also seen in some types of solely deuterically oxidized grains not illustrated in this figure.

3.3.8 ***Silicate minerals.*** Olivine is the silicate mineral most readily decomposed in either deuteric or regional hydrothermal alteration environments. A high degree of deuteric alteration results in marked changes to titanomagnetite, separate ilmenite and olivine. Breakdown of the latter in deuteric conditions has been described by Haggerty & Baker (1967), who report the formation of magnetite or hematite. The regional hydrothermal environment completes the destruction of olivine in basaltic rocks. Thus, in the Zone I lavas of Mull, Bailey *et al.* (1924) report the complete absence of fresh olivine. In some cases at least olivine is replaced by green silicates and chains of small euhedral magnetites. Again, the Zone I Keweenawan lavas examined by us characteristically have olivine replaced either by stringers or nearly complete pseudomorphs of hematite, or by various dull grey to black iron-bearing phases.

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3.4 *Summary of opaque mineralogical changes induced by Regional Hydrothermal Alteration*

Two features deserve mention in a summary:

(i) Very complex mineralogical situations can result from the superposition of spatially variable regional hydrothermal alteration on basalts that have already experienced spatially variable deuteric oxidation.

(ii) The existence of certain alteration products of the primary iron-titanium oxide minerals of basalts may be indicative of minimum reheating temperatures in the regional hydrothermal environment.

In particular, the following relationships appear to be fairly well established on the basis of the secondary mineral stability-field calibration of the zonal scheme (Fig. 1, Table 2).

Ferri-rutile granules in titanomagnetite: a minimum reheating temperature of about 150 °C.

Sphene replacement of ilmenite lamellae in titanomagnetite: a minimum reheating temperature of about 250 °C.

Polycrystalline titanohematite pseudomorphing of titanomagnetite and woven alteration of separate ilmenite: a minimum reheating temperature of about 300 °C.

4. The influence of regional hydrothermal alteration on the strong field thermomagnetic properties of basaltic lavas

4.1 *Introduction*

In this section the strong field thermomagnetic curves of basaltic lavas are examined in order to separate the effects of deuteric oxidation and regional hydrothermal alteration. Regional hydrothermal effects have been looked for initially in strong-field thermomagnetic curves as these curves show properties (Curie points, etc.) which are independent of variables such as amount and grain size of the magnetic phases and the intensity of the original magnetizing field.

The detailed plan for this section is first to look at the effect of different degrees of deuteric oxidation on strong-field thermomagnetic curves, then to turn to the effect of different degrees of regional hydrothermal alteration on specimens in each stage of deuteric oxidation. As shown in Section 3 for opaque petrological properties, combinations of varying degrees of these two alteration processes results in the existence of a great variety of types of thermomagnetic curves. A discussion of the possible palaeomagnetic importance of regional hydrothermal alteration effects is given in Section 5.

4.2 *Method of measurement and definition of parameters*

Strong field thermomagnetic curves were obtained using a modified version of Chevallier & Pierre's (1932) apparatus. This instrument uses a permanent magnet to produce a magnetizing field of about 1000 Oe, which is probably less than the saturation field at ambient temperatures for many basalts. However, actual magnetic moments are likely to be close to saturation moments at ambient temperatures and to be equal to them at moderate temperatures. As described in Ade-Hall, Wilson & Smith (1965) Curie points were taken as being the temperature at which the curvature of the concave part of a heating curve was at a maximum. In addition to conventional Curie points, several other features of strong field thermomagnetic curves were found to be related to the alteration history of specimens:

(i) Initial rise in magnetization on heating, leading to a rounded peak in the curve, is characteristic of certain specimens. This phenomenon was first

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described for basalts by Cox & Doell (1962). We term this feature a hump (see curve for Icelandic specimen H26–1 in Fig. 9).

(ii) Rise in magnetization from a little below 100 °C terminating with a sharp discontinuity in the curve between 100 °C and 200 °C (see curves for Mull specimen C190–1 and Keweenawan specimens D2–5 in Fig. 9). We term this feature a kink.

(iii) Frequently in specimens containing the kink feature an irreversible inflexion in the heating curve occurs between 300 °C and 450 °C. (Figs 9 and 18.) It is not clear whether this is a true Curie point or represents some chemical change in the magnetic phases of specimens showing this feature. To separate this feature from conventional Curie points we have identified it by an ‘ e ’ in Curie point histograms.

4.3 *Thermomagnetic curves for different degrees of deuteric oxidation*

Unaltered basalts, with magnetite oxidation numbers (Section 3.2) close to unity and no indications of regional hydrothermal alteration, frequently have simple strong field thermomagnetic curves with single, low (0–200 °C) Curie points. Réunion specimen RU95–C3 of Fig. 9 illustrates this condition. Electron microprobe analysis of the titanomagnetites of these specimens yields Fe : Ti ratios consistent with these low Curie points. A fraction of specimens, *apparently* in this petrological condition do have single high Curie points. The reason for this behaviour is not yet known but could be due to rather strong *local* hydrothermal alteration during the later stages of lava cooling.

The advent of moderate deuteric oxidation in the more typical, single low Curie

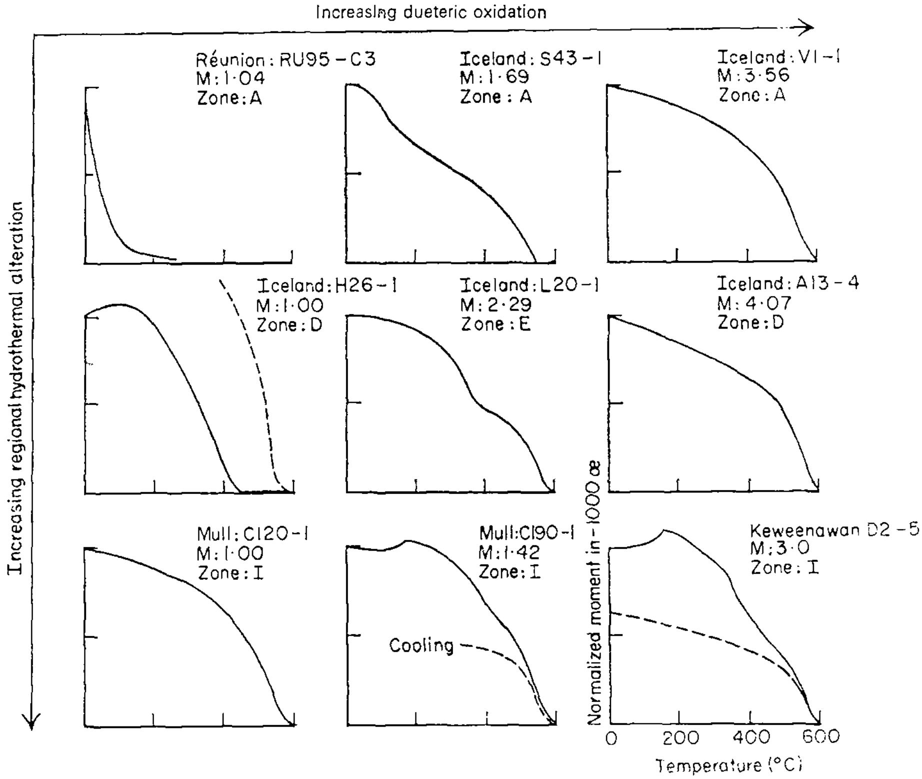

FIG. 9. The evolution of strong field thermomagnetic curves.

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point basalt, results in the formation of material with a second, high, Curie point, often between 550 °C and 600 °C, that is close to that reported for pure magnetite. (Iceland specimen S43–1 of Fig. 9). This thermomagnetic behaviour is consistent with microscopic observation. Specimens with an *average* moderate state of deuteric oxidation characteristically contain individual grains both in low and higher states of deuteric oxidation. The grains without deuteric oxidation must be responsible for the low Curie point while the newly-formed, titanium deficient magnetite of grains containing ilmenite lamellae, will be responsible for the high Curie point. We refer to this type of curve as a double Curie point curve. The greater specific magnetization of the titanium-free magnetite of the exsolved grains results in the appearance of a high Curie component after only a slight degree of deuteric oxidation.

Further deuteric oxidation results in an increase in the proportion of the contribution of the high Curie point material to the total magnetization, with little if any change in either Curie points, until, when the average magnetite oxidation number reaches about 3·3, the contribution of the low Curie point phase is no longer discernable (Icelancid specimen V1–1 of Fig. 9). Again, this is consistent with microscope observations; specimens with magnetite oxidation numbers in excess of 3·3 usually contain few if any non-deuterically oxidized grains. The type of curve produced has a characteristically smooth geniculate form and is termed a single high Curie point curve.

Thus, a typically patchily deuterically oxidized (Section 3.2) unburied basalt flow will yield, from different parts of the flow, thermomagnetic curves of the single low, double and single high Curie point type.

4.4 *Thermomagnetic curves for different degrees of regional hydrothermal alteration*

In Section 4.3 we pointed out that the low Curie points of unburied lavas were consistent with estimates based on the composition of homogeneous titanomagnetites obtained from electron microprobe measurements (Fig. 10). However, many other basalts, seen with the microscope to contain homogeneous titanomagnetites, and with low probe-determined Fe : Ti ratios, have anomalously high Curie points compared with values for stoichiometric titanomagnetite (Ade-Hall 1964; Smith 1967a) while others have Curie points agreeing well with estimates based on microprobe measurements (Carmichael 1965; Creer & Petersen 1969). At the time these

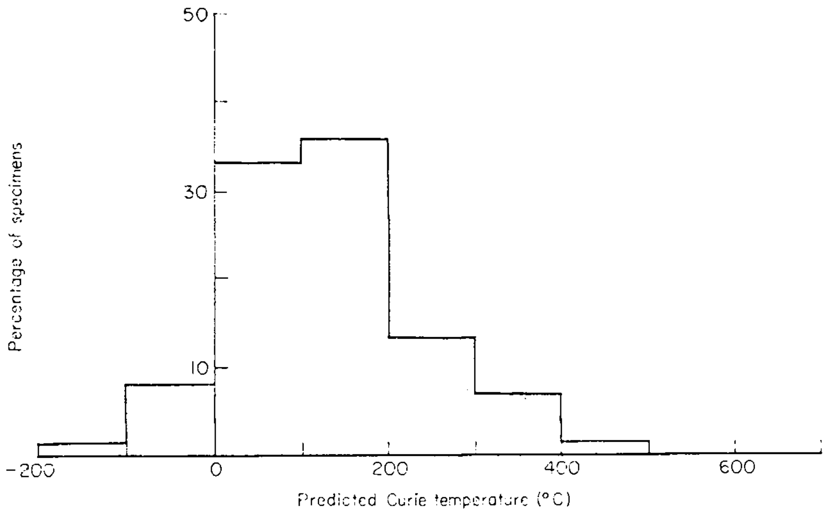

FIG. 10. Curie points predicted from electron probe microanalyser results for 75 non deuterically oxidized basalts.

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results became available it was not possible to explain the pattern of agreement and disagreement between calculated and measured Curie points. However, it is now possible at least largely to explain this spread of Curie points in terms of degree of regional hydrothermal alteration. Thus, historic, and therefore unburied and at least largely unaltered lavas, tend to have strongly bimodal Curie point distributions (Fig. 11). For these flows the low Curie points are in the 0–300 °C range, with most being between 100 and 200 °C. This last temperature interval also contains the biggest single percentage of microprobe-measurement-based estimated Curie points (Fig. 10). Thus it is likely that the optically homogeneous titanomagnetite of these historic basalt flows approximates in composition to stoichiometric high titanium titanomagnetite. The high Curie points in the historic flow distributions probably at least largely represent the results of varying degrees of deuteric oxidation in the flows. Division of the Curie points of specimens from 213 Eastern Icelandic lavas spaced equally through a collection of about 1100 flows (Dagley *et al.* 1967), into regional hydrothermal alteration zone groups (Fig. 12) allows us to examine the effect of

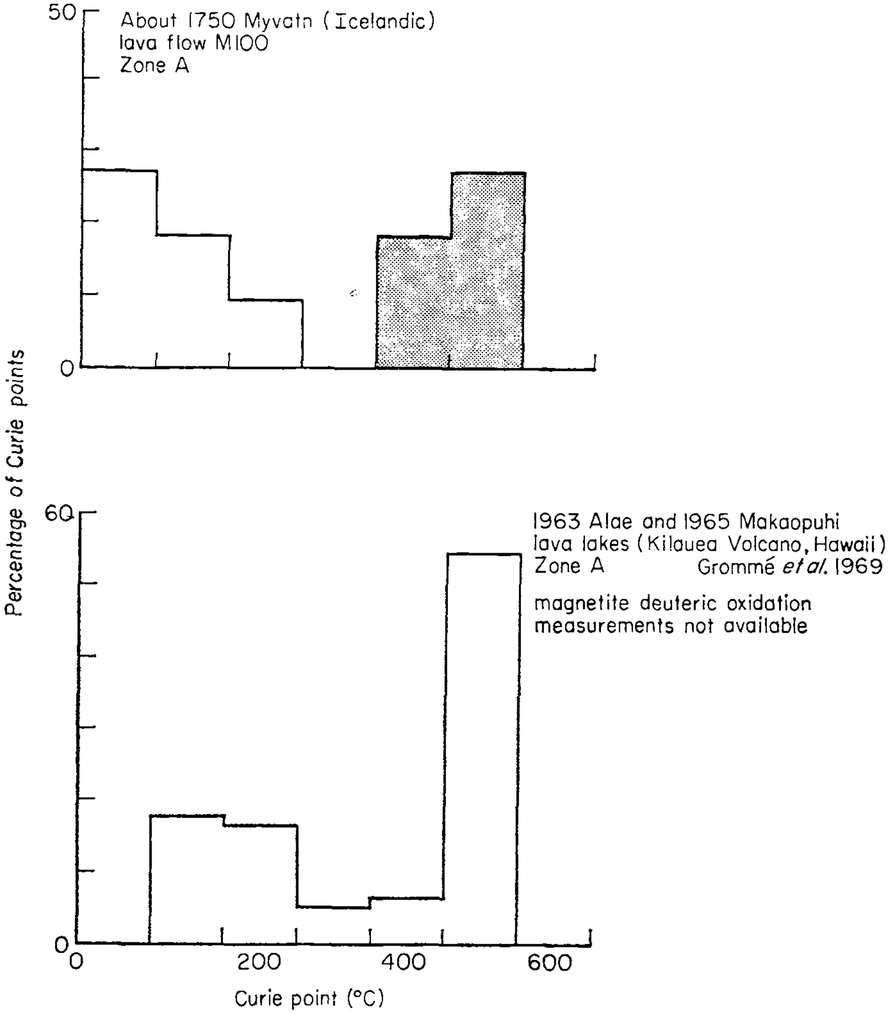

FIG. 11. Curie point distributions for historic basalt lavas. Stippled portion of upper histogram indicates Curie point results from deuterically oxidized specimens.

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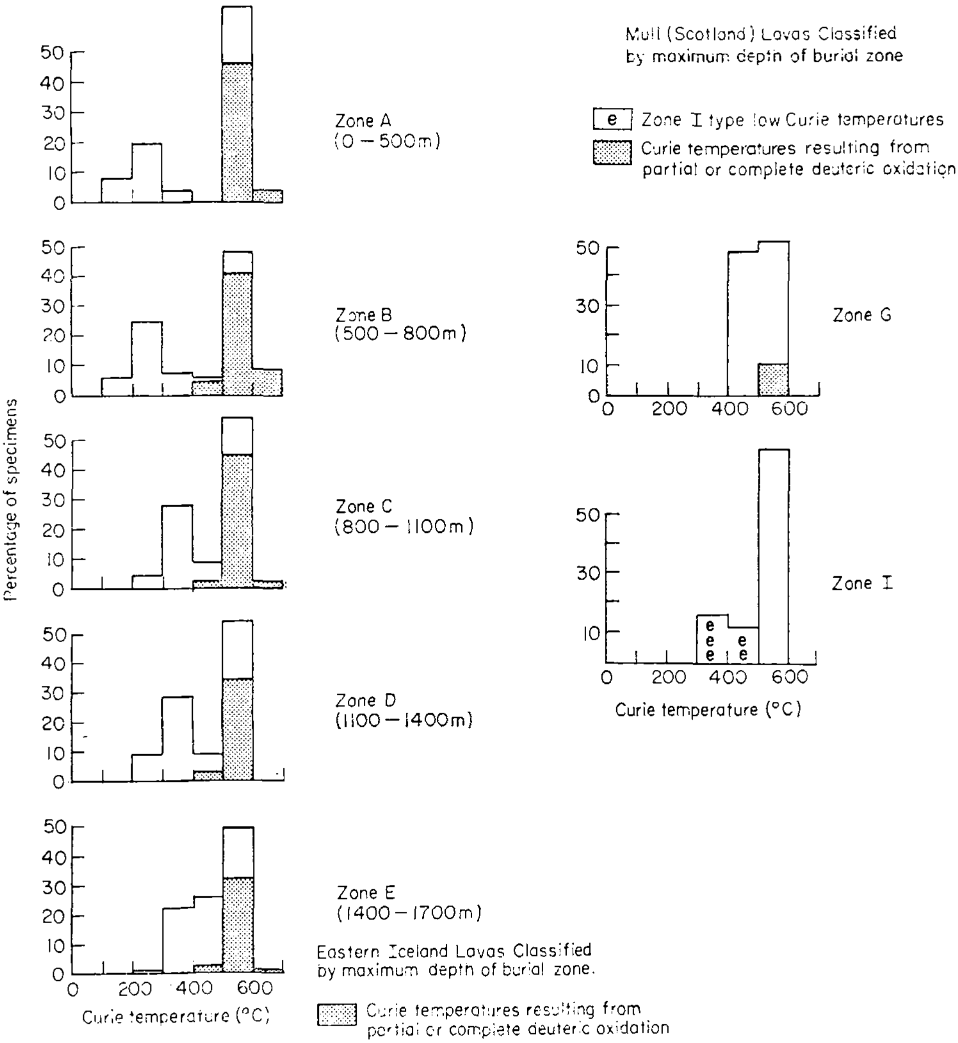

FIG. 12. Curie point distributions for lavas from the regional secondary mineral zones of Eastern Iceland and Mull, Scotland.

increasing alteration on the strongly bimodal distribution found for historic lavas. We see that the largely zeolite free lavas of zone A also show a strongly bimodal Curie point distribution, with a slightly higher mean temperature for the low Curie point peak than in the historic lavas. Increasing burial, resulting in increasing degree of regional hydrothermal alteration, causes a gradual migration of the low Curie peak to higher temperatures through zones B, C and D until with zone E degree of alteration the low Curie point peak cannot be resolved from the high Curie point peak, this latter largely representing deuterically oxidized titanomagnetite, which has remained in the 500–600 °C interval throughout. Our Icelandic collection does not contain material that has experienced regional hydrothermal alteration to greater than Zone E conditions, and, as our Keweenawan collection is entirely from Zone I we turn to the Mull (Scotland) lavas to continue the trend shown by the Icelandic lavas. The Mull lava collection represents both the regionally hydrothermal altered Gribun lavas of Zone G and the Zone I 'pneumatolysed' lavas surrounding the Mull intrusive centre (Walker 1970). The Zone I lavas are in fact younger than the

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Zone G lavas. We see a quite sharp single peak for the Zone G Curie points—notwithstanding the fact that the specimens measured were all in a low state of deuteric oxidation *all* the Curie points between 400 and 600 °C. Finally some of the Zone I thermomagnetic curves have single high Curie points while others show the complex highly irreversible type of thermomagnetic curve containing the kink feature, etc. Illustrated in Figs 9 (specimens C190–1, D2–5) and 18. This represents an entirely different kind of behaviour, as distinct from the gradual migration of conventional Curie points occurring from Zones A to G. Unfortunately our Mull lava collection contains no specimens from Zone H, but it seems from lavas from other localities that this new type of thermomagnetic behaviour first occurs after alteration to Zone H conditions.

We mention at this point that several lines of evidence show that we are dealing with burial induced changes rather than time dependent changes in magnetic properties. In the Eastern Icelandic lava succession, low Curie points (for Zone A lavas) occur in two positions in our succession, among the youngest flows and *again* at about two-thirds of the way towards the oldest flows. These two groups of little buried flows are separated by several thick sequences of flows that have experienced D and E zone conditions. The stratigraphically old but little buried lavas probably belong to the ‘ Grey Stage ’ lavas of Pjetursson (1910) which were originally thought to be a disconformable younger sequence but are now seen to owe their pale coloration and empty vesicles to the absence of regional hydrothermal alteration effects (Walker 1960). Again, the basaltic rocks from the different parts of the British Tertiary volcanic province, which are probably rather closely grouped in age within the Palaeogene, show widely different Curie point distributions (Fig. 13). Thus, the

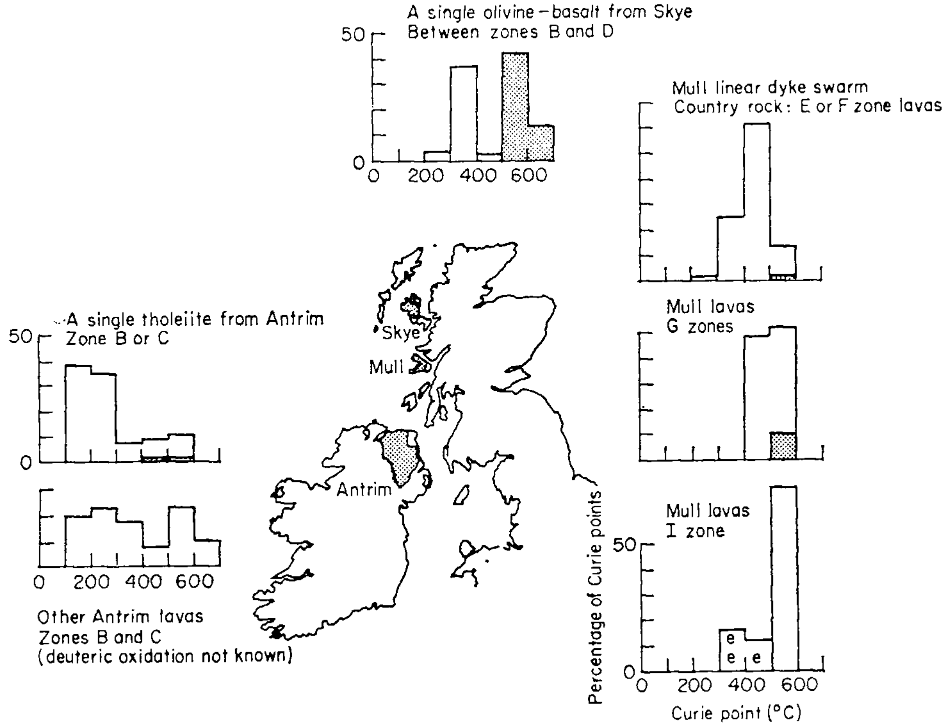

FIG. 13. Curie point distributions for basalt lavas and dykes from different parts of the British Tertiary Volcanic Province. Stippled portions of histograms indicate Curie point results from deuterically oxidized specimens.

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Antrim lavas, with zeolite minerals indicating zone B or C regional hydrothermal alteration conditions have a marked bimodal Curie point distribution with Curie points down to 100 °C. In contrast, Curie points for a single lava from Skye show a 300–400 °C low Curie point peak, and *all* the Mull material shows unimodal distributions corresponding to the most extreme regional hydrothermal conditions known in the province.

While the case for Curie point changes being burial and not time dependent seems quite conclusive we must point out that the *probability* of a basalt unit having experienced regional hydrothermal alteration must increase with its age because the chances of burial, due to orogenic or epeirogenic processes, must increase with time. For example, while many Plio-Pleistocene lavas have Curie points in the 100–200 °C interval, a rather smaller proportion of Paleogene lavas have such low Curie points, and no Carboniferous or Devonian lava known to us has a Curie point of less than 300 °C. This increased probability of regional hydrothermal alteration with age must be the explanation of the relatively small number of low Curie points found for basalts. The fact that there are two alternative paths to single high Curie points curves accounts for this (Fig. 9). This figure shows that either high deuteric oxidation *or* high regional hydrothermal alteration can produce single high Curie point curves (specimens V1–1 and C120–1). Clearly, selection of material for special studies, for example, ancient field intensity determinations for which high deuteric oxidation material may be required cannot be made from strong field thermomagnetic curves alone. This strong field thermomagnetic curve evolution diagram shows several other points of interest:

(i) Intermediate Curie point curves characterized by a hump and a great increase in strong field moment and Curie point after heating to 600 °C are characteristic of very low deuteric oxidation basalts that have experienced moderate regional hydrothermal alteration conditions. The hump is not suppressed by increased field at least up to 2200 Oe (Cox & Doell 1962). This type of curve is particularly common for submarine lavas (Cox & Doell 1962; Ade-Hall 1964; Ozima, Ozima & Kaneoka 1968) which are characteristically in a very low state of deuteric oxidation (Ade-Hall 1964; Haggerty 1970; Watkins, Paster & Ade-Hall 1970). However a number of continental basaltic lavas and dykes also have this type of opaque mineralogy and corresponding strong field thermomagnetic properties. The importance of the place of the magnetism of submarine lavas in the synthesis of the new global tectonics makes it imperative to understand the unusual strong field thermomagnetic behaviour of this type of material.

(ii) Double Curie point curves for specimens that have experienced intermediate regional hydrothermal alteration very rarely show humps. This, even a little ilmenite exsolution from magnetite leaves a material that, after moderate hydrothermal alteration, is relatively little changed during laboratory thermal cycling. For a lava that experienced variable deuteric oxidation, all low Curie points will rise by approximately the same temperature on regional hydrothermal alteration and the ratio of contributions of high and low Curie point components to the total strong field magnetization will still be closely related to the degree of original deuteric oxidation (Ade-Hall *et al.* 1968).

(iii) When I (or possibly H) zone degree of alteration has been achieved a wholly new type of thermomagnetic curve occurs, containing both the kink feature and the ‘ e ’ type Curie point. Two points are relevant concerning this new type of thermomagnetic behaviour. Firstly, it seems to show a weak preference for occurrence in originally high deuteric oxidation specimens, although this is not so clear cut as is suggested in Fig. 9. Secondly, the fact that

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originally high deuteric oxidation specimens, which appear to have reached Zone G conditions without apparent magnetic alteration, are now showing some changes in magnetic properties, must make their use for use in ancient field intensity determinations (Smith 1967b) open to question.

4.5 *Comparison of results from other basalt areas where regional secondary mineral zones have been identified*

In this section we compare results from a number of other basalt sequences, etc., for which both regional hydrothermal alteration conditions and thermomagnetic behaviour are known, with the general scheme of the evolution of thermomagnetic curves described in the proceeding section.

4.5.1 *Three single lava studies.* One of the most puzzling results of several detailed studies of single lava flows (Wilson *et al.* 1968; Ade-Hall *et al.* 1968; Lawley & Ade-Hall 1970) concerned the strong field Curie point distributions. The problem was to explain the fact that while the flows all had similar magnetite deuteric oxidation ranges ($1 \cdot 0 < M < 4$) each flow had a different characteristic Curie point distribution

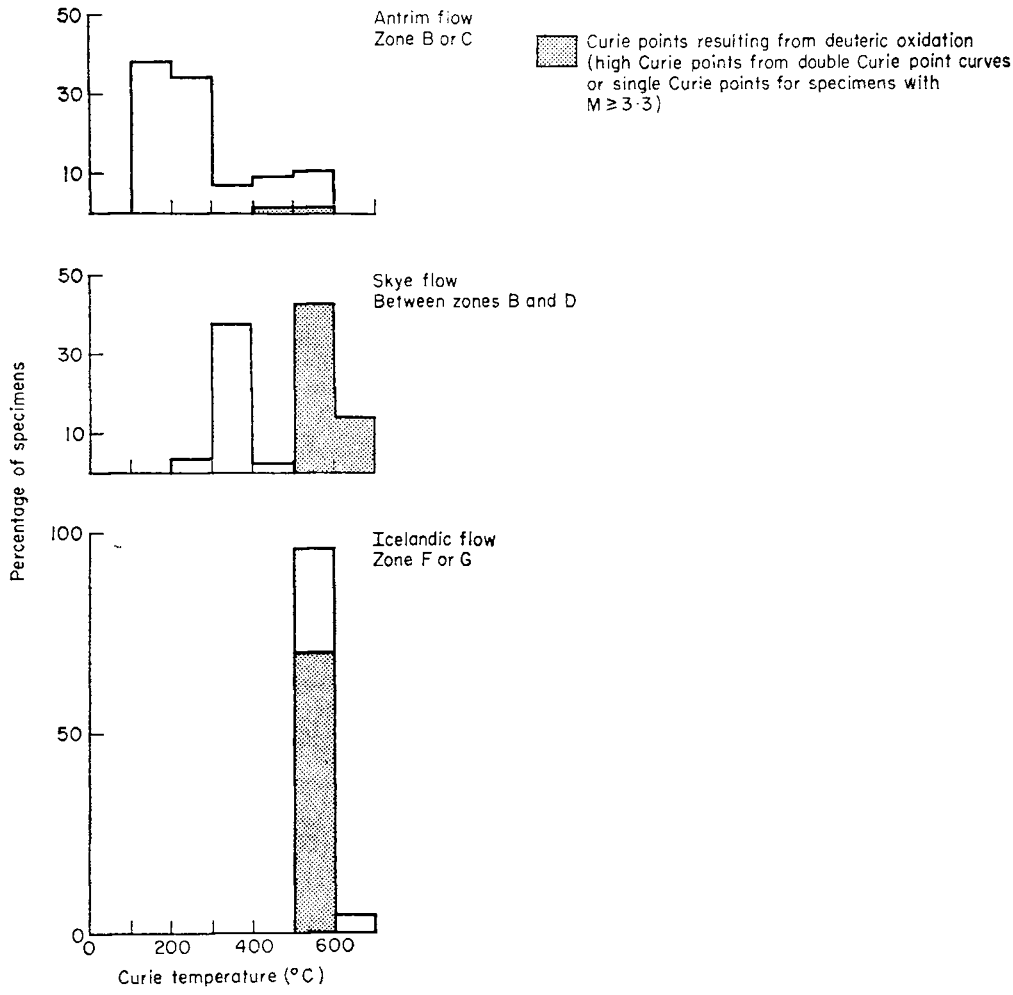

Fig. 14. Curie point distributions from three detailed studies of single basalt flows.

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(Fig. 14). The explanation of these different distributions appears to lie in the different regional hydrothermal alteration histories of the flows. The Antrim tholeiite flow (Lawley & Ade-Hall 1970) has been little buried and the Icelandic flow of Wilson *et al.* was sampled near to the edge of a previously identified zone of propylitization which probably surrounds a volcanic centre. It is interesting that the Icelandic flow showed a fairly constant whole rock $Fe_2O_3/(Fe_2O_3+FeO)$ ratio throughout. This fact is consistent with a highly cation-deficient nature for the low deuteric oxidation titanomagnetites, so hinting at a solution to the problem of the nature of the mineralogical change to titanomagnetite expressed by a gradual Curie point rise.

4.5.2 *The British Tertiary volcanic province.* As mentioned in Section 4.4 and illustrated in Fig. 13 the Curie point distributions for basaltic lavas and dykes from different parts of this province are consistent with their regional hydrothermal alteration history. A point of interest concerns the time of alteration of basaltic dykes. The Tertiary dykes of Mull, Scotland ($1\cdot0 \leqslant M \leqslant 2\cdot0$) are now exposed at a level that may well have been quite deep in the lava pile at the time of intrusion.

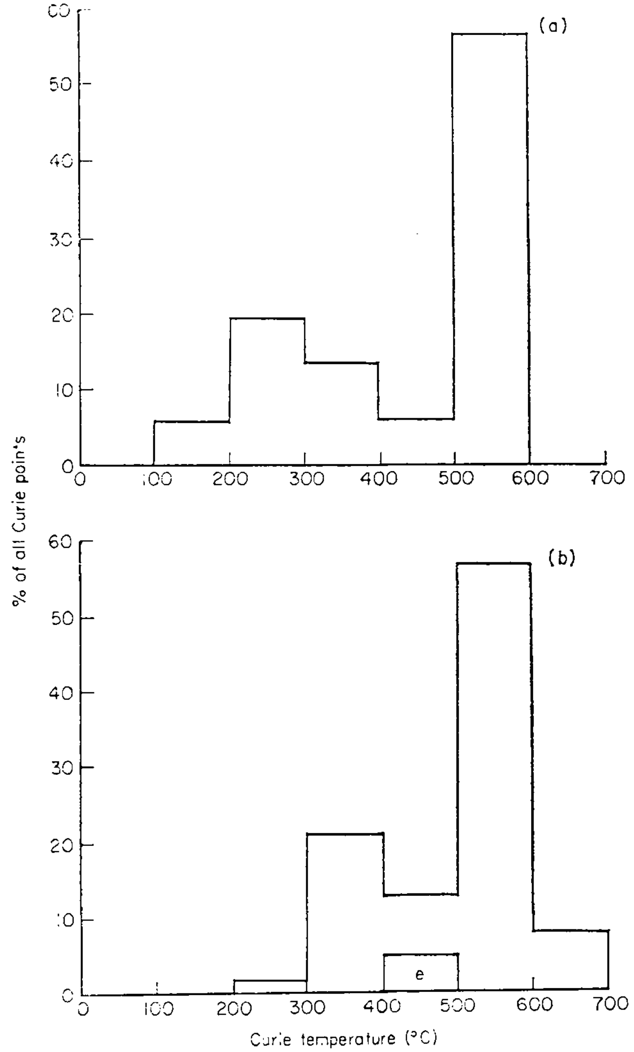

Fig. 15. Curie point distributions for the Simien (Ethiopian) lavas.

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Thus, hydrothermal alteration of the dykes in these circumstances could have taken place very soon after intrusion in contrast with the necessary delay in the case of lavas.

4.5.3 *The Simien (Ethiopia) lava pile.* This area is particulary interesting from the point of view of regional hydrothermal alteration as a sequence of 139 flows is exposed in a single vertical section more than 2 km high. A proper study of the secondary minerals in these lavas has yet to be made but examination of the drill cores taken for palaeomagnetic work allows recognition of a simple zonation. The 69 lavas comprising the top 970 m of the section are free of secondary minerals while the remaining stratigraphically lower flows are characteristically zeolitized. Curie point histograms for the zeolite free and zeolite bearing zones are shown in Fig. 15. The higher values (300–400°C interval) of the low Curie point peak for the more deeply buried lavas (Fig. 15(b)) is consistent with the relationship between Curie point and burial found elsewhere. Two other points of interest arise from the study of the Simien lavas. The combination of a thick zeolite free zone and low temperature low Curie point peaks compared with the situation in Eastern Iceland suggests a lower temperature gradient during lava pile formation in Simien region. Again, the *occasional* occurrences of thermomagnetic curves with kink features and ' e ' type Curie points within the zeolite bearing zone, elsewhere characteristic of H or I conditions is surprising and requires further investigation.

4.5.4 *The Deccan (Indian) lava pile.* Recent work by Walker (personal communication) has resulted in the identification of regional secondary mineral zones in this major plateau lava area. In general, the lavas of the western (Bombay) side of the area have been more deeply buried than those on the eastern (Nagpur–Hyderabad) side. We were fortunate in having access to an early palaeomagnetic collection from the Deccan Traps (Clegg, Deutsch & Griffiths 1956) where two sampling areas were located in different regional secondary mineral zones. Curie points histograms for small collections from each of these two areas are shown in Fig. 16.

Comparison of these histograms shows that the low Curie point interval for the Linga lavas 16(a), from the eastern side of the lava area, is significantly lower than that for the Khandala area, 16(b) which is near to Bombay. In this semiquantitative sense the Deccan data agrees with the results from the standard successions. However, in detail there is some apparent conflict. Thus, the Curie point results for the Linga specimens suggest that regional secondary mineral zones from A to C may be present, while the field study of zeolite occurrences indicates that Zone C is dominant in the area. Again, the Khandala magnetic results suggests that Zones C or D and H or I are present whereas the field work indicates that Zones D or E are dominant in the area. These apparent conflicts might well be explained if detailed studies of the Deccan Trap secondary mineral assemblages for the relevant areas were available.

4.5.5 *Jurassic tholeiites of the Central Transantarctic Mountains.* It has been possible to examine a series of generally low deuteric oxidation Gondwana tholeiites for which detailed knowledge of the secondary mineral content is available. The secondary minerals present strongly suggest a zone E/F degree of regional hydrothermal alteration (Elliot, in press) and this classification is supported by the presence of granulation in the titanomagnetites of several flows. Fig. 17 shows a histogram of Curie points. With or without the inclusion of two specimens, yielding curves with ' e ' type low Curie points (and kinks) the distribution of low Curie points closely matches the Zone E distribution for the Eastern Icelandic lavas (Fig. 12). Specimens from a tholeiiteic sill within the Beacon sequence some 2000 m stratigraphically below the base of the lavas contain exsolved titanomagnetites with sphene replacement of ilmenite lamellae; these specimens have thermomagnetic curves with kinks and ' e ' type low Curie points.

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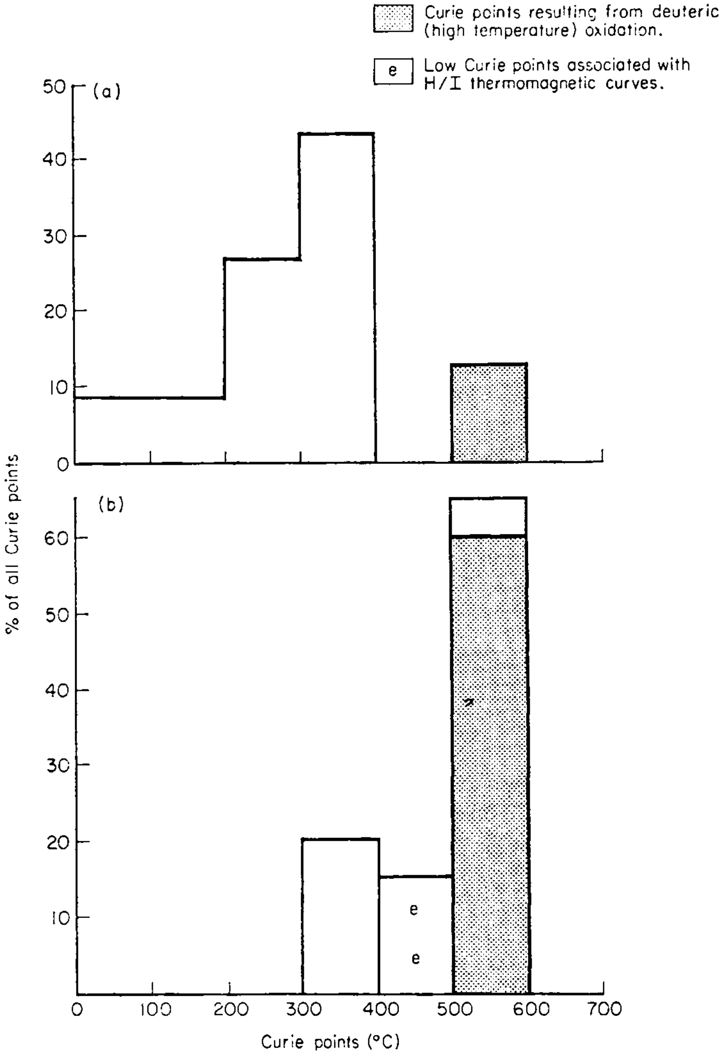

FIG. 16. Curie point distributions for lavas from two areas in the Deccan Traps, India.

4.5.6 *The wide distribution of Zone H/I characteristic thermomagnetic curves.* The geographically and geologically wide distributions of the striking thermomagnetic curves found for many basalts that have experienced Zone H or Zone I alteration conditions is impressive. Fig. 18 illustrates this type of curve, with its characteristics redefined, and shows Curie point histograms from four lava series, each containing a significant fraction of this type of curve. Note that wide geographic spread of locations, the wide age range (Tertiary–Carboniferous–Pre-Cambrian) and that Peruvian andesites yield curves apparently identical with those found for basalts. This type of thermomagnetic behaviour has also been found for Cretaceous andesites from Chile (Thompson, personal communication), submarine basalts (Section 4.6), lavas contributing to the time polarity scheme (Section 4.6) and rarely among the Ethiopian, Indian and Transantarctic Mountains lava sequences described in Sections 4.5.3, 4.5.4 and 4.5.5 respectively.

Summarizing the results from the various areas described in this section we can see that there is agreement, at the present level of knowledge, between the results

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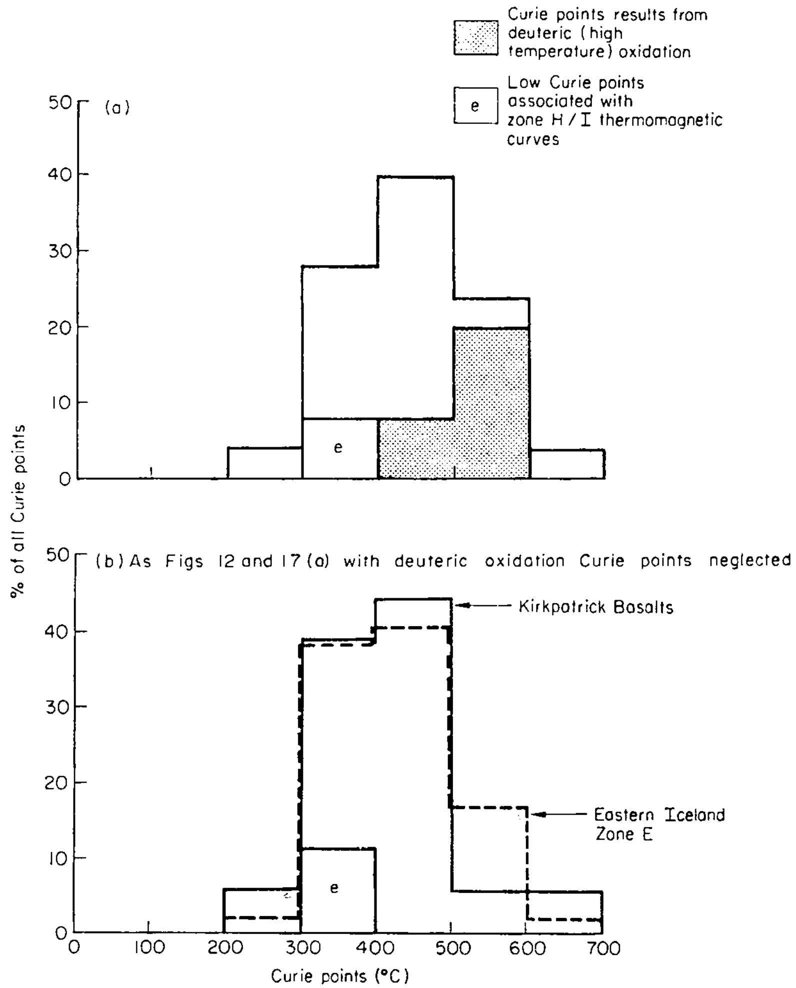

FIG. 17. Curie point distribution for the Kirkpatrick Basalts of the Mid-Antarctic Mountains.

from these areas and those for the standard successions described in Section 4.4. We propose that the relationships described here between thermomagnetic properties and regional hydrothermal alteration are probably characteristic and where lava secondary mineral zonation is unknown, the strong field thermomagnetic and opaque petrological properties may be used to estimate the regional hydrothermal alteration history.

4.6 *Strong field and opaque petrological characteristics of basalt lavas from other areas*

In this section we use these properties to try to reconstruct the regional hydrothermal history of various basalt lavas.

4.6.1 *Submarine lavas.* This type of material, with its characteristic low deuteric oxidation state, is ideal for the study of regional hydrothermal alteration induced changes in thermomagnetic properties.

There are already several reports in the literature of submarine lavas exhibiting zeolite facies metamorphism (Aumento & Loncarevic 1969 and others) and also of greenschist facies metamorphism (Matthews, Vine & Cann 1965; Melson & Van

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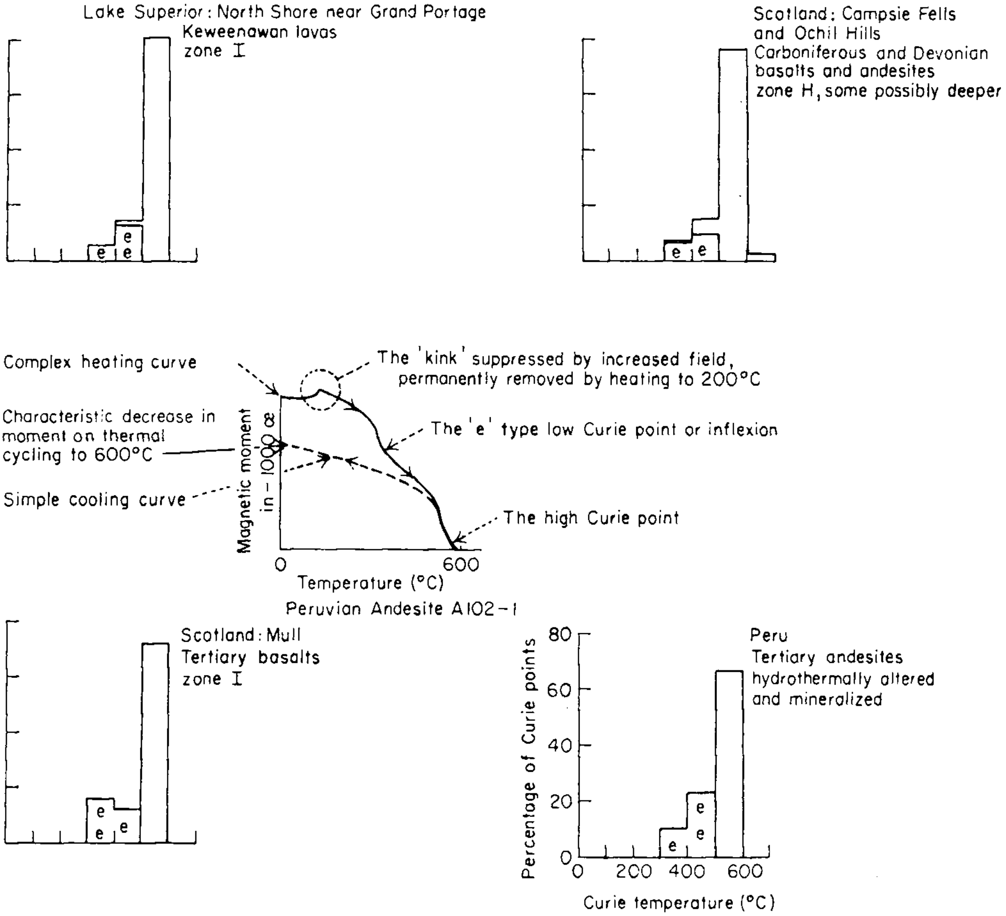

FIG. 18. The characteristic H/I zone strong field thermomagnetic curve and its widespread age and geographical distributions.

Andel 1966 and others). Thus we may expect corresponding types of thermomagnetic curves, and these in fact, have been found to occur. Fig. 19(a) shows Curie point histograms for collections from dredge stations close to the escarpment of the Mendocino fracture zone. Both collections contain specimens with Curie points ranging from 260 °C to 500 °C. This spread in Curie points, and the variation of thermomagnetic behaviour in general is consistent with sampling from a wide range of secondary mineral zones. The range could have encompassed Zones C to I. The discovery of basalt specimens from a wide range of zones in collections taken near a major fault or scarp is not surprising, but the geographical variation of Curie points found in the North Atlantic (Fig. 19(b)) is quite unexpected. In this figure, Curie points for 45 submarine lava specimens either recorded in the literature or determined by ourselves have been arranged in groups dependent on their distance from the Mid-Atlantic Ridge. It is immediately obvious that a systematic variation of average Curie point with distance from the ridge occurs, with low (100–200 °C) Curie points largely confined to within 100 km of the ridge axis, and as yet unrecorded at further than 24 km from the axis. Increase in the average Curie point is most rapid within 100 km from the ridge axis, a fact already noted by Schaeffer & Schwarz (1970) whose data has been included in our analysis and is consistent with other results. This

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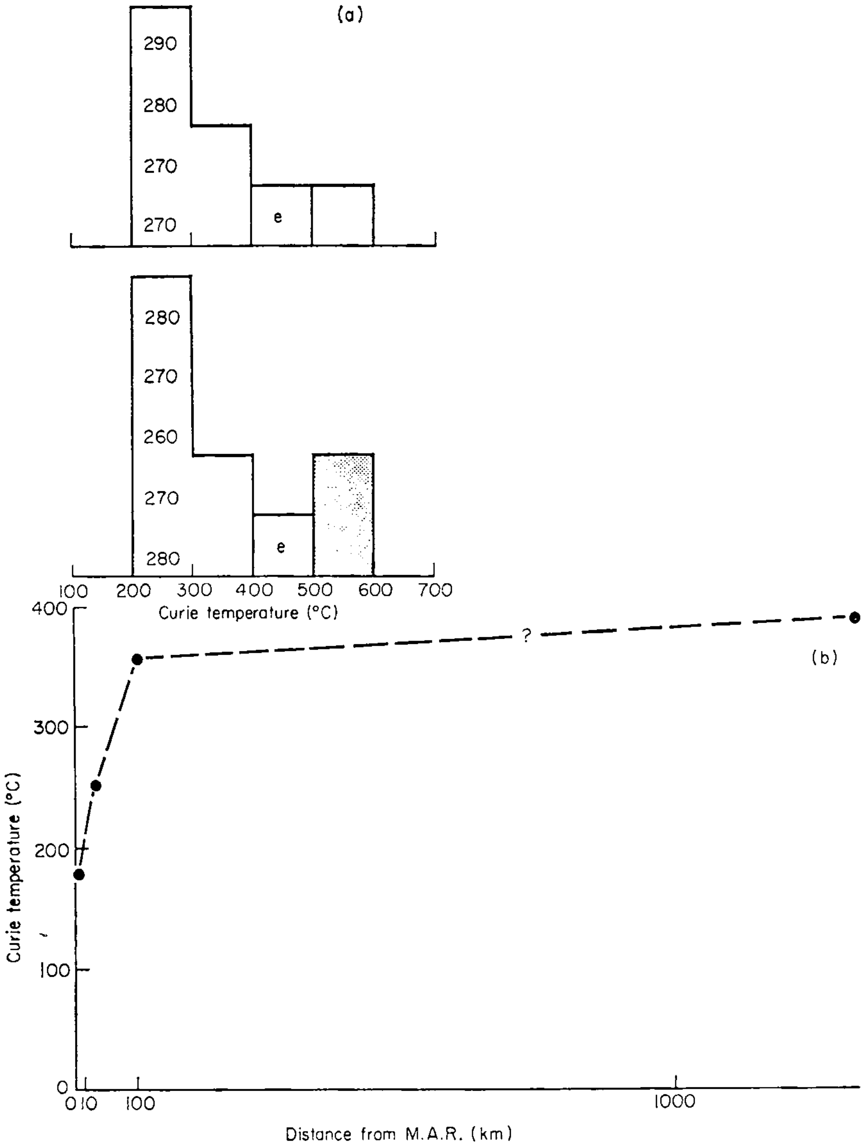

FIG. 19. (a) Curie point distributions for specimens dredged from the Mendocino scarp area. Upper: Dredge FAN BD 33. Lower: Dredge FAN BD 25. (b) The rise in submarine basalt Curie points away from the Mid-Atlantic Ridge (0–10 km interval; 13 Curie points, 10–100 km interval; 19 Curie points, 100–1000 km interval; 5 Curie points, >1000 km; 8 Curie points).

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increase in average Curie point is matched by an order of magnitude decrease in natural remanence intensity and an increase in the Fe_2O_3/FeO ratio (Irving 1970). The paucity of results for specimens in the 100–1000 km interval means that extrapolation is in doubt here but results at >1000 km appear to confirm the trend. It is not clear whether seamount data should be included in the analysis. This question arises especially in connection with the small group of Curie points for specimens at >1000 km from the M.A.R. axis. We have included two values from seamount material, one at 210 °C for a specimen of basalt with unfilled vesicles from a seamount 250 km north of Maderia (Laughton, Hill & Allan 1960) and the other at 370 °C from the Discovery Seamount. Exclusion of these two values raises the average Curie point for this interval to 420 °C.

Thus it seems that the regional hydrothermal alteration of the exposed submarine basalts increases both to the east and to the west of the Mid-Atlantic Ridge in the North Atlantic. It must be asked why such a relationship exists. A major uncertainty in answering this query is the lack of knowledge of the mechanisms and rate for erosion of basalts on the floor of the deep oceans. We suggest that either an unrecognized erosional process acting on exposed basalt on the ocean floor is capable of removing hundreds of metres or even a few kilometres of basalt in a time interval of the order of 100 M.a. (Mega année), or that chemical reaction between basalt and sea water at about 0 °C results in changes in the basalt similar to those produced on the continents by regional hydrothermal alteration.

A similar analysis has been attempted for the Curie points of Pacific submarine basalts. Less information is available than for the Atlantic and values for specimens close to and at known distances from the East Pacific Rise have yet to be determined. However, widely-scattered specimens, many at great distances from the East Pacific Rise, have lower Curie points than predicted from the North Atlantic model (Ozima *et al.* 1968; Watkins, Paster & Ade-Hall 1970). The significance of seamount material must again be questioned. Active volcanicity well away from the Rise, such as in the Hawaiian Islands and elsewhere, introduces proportionally low Curie point values if included with values for basalts generated on the Rise and carried to their present positions by sea-floor spreading.

4.6.2 *Lavas contributing to the time-magnetic polarity scale.* We have been able to examine the opaque petrology and strong field thermomagnetic properties of a number of specimens from basalt flows contributing to the time-magnetic polarity scale of Cox, Doell & Dalrymple (1968). A Curie point histogram and age-Curie point plot are shown in Fig. 20. Since all the lavas are <4·5 M.a. old we can expect many examples of little or no post-eruption burial, and this is indeed evident from the strongly bimodal nature of the Curie point histogram. However, a significant number of specimens (6, or 29 per cent of the total) do show evidence of considerable hydrothermal alteration, which of course may not always be of regional burial origin. Four of these specimens come from California and the other two from the Pribilof Islands. Five of these six specimens have single high Curie points in the 480–560 °C interval, suggesting perhaps Zone G maximum burial conditions, while the sixth has the characteristic H/I zone curve, complete with kink, 'e' type Curie point and considerable decrease in moment on cooling. Thus the first five specimens may have been reheated to temperatures between 200 and 300 °C and the sixth in excess of 300 °C. Two questions must be asked concerning these six specimens; whether their natural remanence polarity or their potassium or argon contents could have been affected by reheating to these degrees at some time possibly considerably after eruption. General discussion of these questions will be kept for Section 5 but we can make a test of the possibilities here by examining the situation of the flows corresponding to these six specimens in the time magnetic-polarity scale itself. For example, if each of these specimens lay in the middle of a long sequence of flows of

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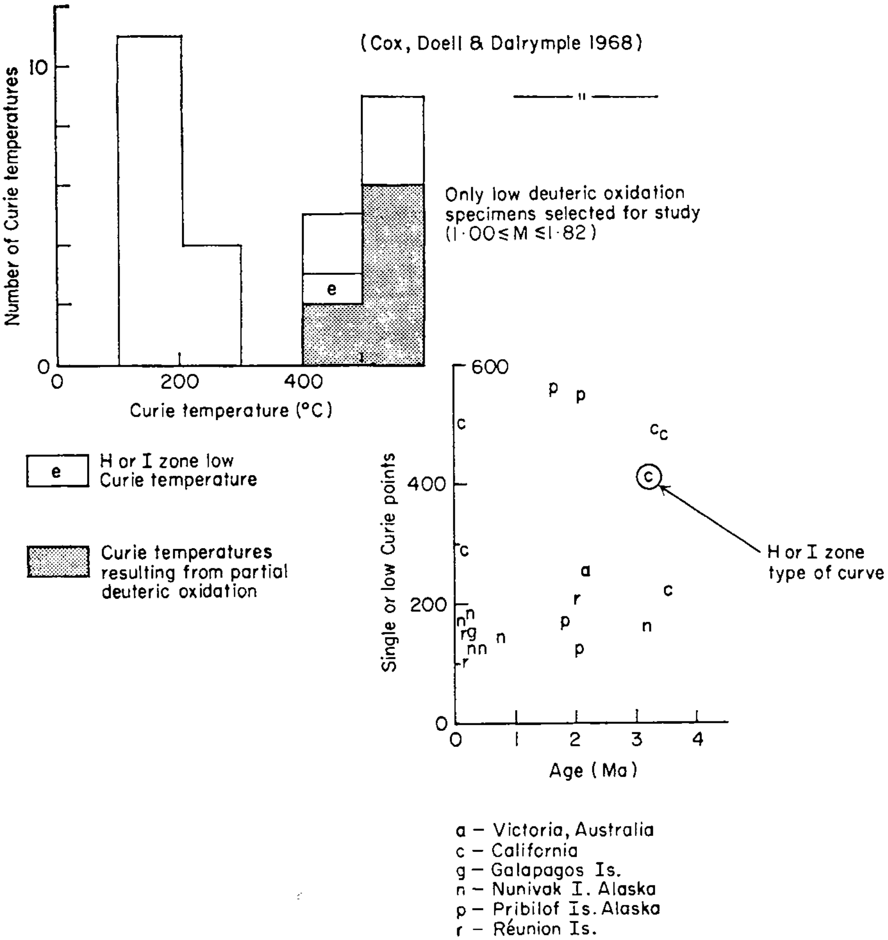

FIG. 20. Curie point distribution and Curie point age plot for basalt specimens from flows contributing to the world wide time-magnetic polarity scale (Cox, Doell & Dalrymple 1968).

the *opposite* polarity there would be grounds to doubt either or both the apparent ages or polarities of the specimens. Referring to Table 1 of Cox *et al.* (1968), the six specimens in question correspond to code numbers 3, 78, 103, 123, 128 and 135. No overall picture emerges of the relationship of the six flows in question to adjacent flows. Flows 3 and 135 occur well within long sequences of one polarity, normal and reverse respectively, so suggesting that both polarity and age for these flows are reasonable. Flows 78, 103, 123 and 128 all occur close to polarity transitions or events, which may or may not have been fixed with certainty to date.

5. Regional hydrothermal alteration and the palaeomagnetism and potassium–argon dating of basaltic rocks

In this section we consider the possible answers to three questions arising from the effects of regional hydrothermal alteration described in the preceding three sections. These questions are:

(1) What are the natures of the magnetic phases produced by the regional hydrothermal alteration of basalts?

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(2) What is the effect on the original thermoremanent magnetization of basaltic rocks of these changes in magnetic minerals?

(3) Can the regional hydrothermal alteration of basalts affect properties relevant to potassium–argon dating?

5.1 *New magnetic phases produced as a result of the regional hydrothermal alteration of basalts*

Strong field thermomagnetic tests suggest that at least two new magnetic phases are produced as a result of the regional hydrothermal alteration of basalts. One phase is that derived from the original high titanium titanomagnetite of unburied lavas and is characterized by a single Curie point higher than that of the original titanomagnetite. The other phase, or phases, is found in the more highly altered basalts and is characterized by the complex curve shown in Fig. 18. The evidence for the nature of these two phases is given separately.

5.1.1 *The high single Curie point magnetic phase of moderately buried basalts.* This phase has not been identified with any degree of certainty but several lines of evidence describe characteristics. The discussion can be formulated in two steps, first, a decision on whether we are dealing with a single or multiphase material, and then the nature of the phase (or phases) will be described.

The widespread recognition of deuteric oxidation in basalts, resulting in the separation of original basaltic titanomagnetites into two or more phases, has prompted the suggestion that exsolution on a sub-microscopic scale might also occur, notwithstanding the absence of visible separation at a magnification of × 1200. Indeed Petersen (1962) and Meitzner (1963) have shown this to be the case in some naturally occurring magnetites. However, three lines of evidence suggest that this is *not* the general mechanism for the formation of the single high Curie point magnetic phase. Firstly, Curie points below room temperature corresponding to an (exsolved) rhombohedral phase are generally absent (Ade-Hall *et al.* 1965). The second line of evidence is related to the first; the stability of natural remanent magnetization (Wilson *et al.* 1968), which is very sensitive to the development of an exsolved rhombohedral phase in magnetite (Ade-Hall 1969), is at least largely independent of the degree of regional hydrothermal alteration (Table 3).

Table 3

Stability of natural remanence (Wilson et al. *1968) for low deuteric oxidation specimens* $(1 \cdot 0 \leqslant M \leqslant 1 \cdot 20)$ *classified on the basis of regional secondary mineral zones*

Eastern Iceland lavas	Number of specimens	$S^*_{200} \pm \Delta S_{200}$	Temperature interval containing low Curie point peak
Zone A	5	0·41 ± 0·11	200–300 °C
B	16	0·29 ± 0·04	200–300 °C
C	10	0·23 ± 0·05	300–400 °C
D	18	0·24 ± 0·04	300–400 °C
E	18	0·27 ± 0·04	400–500 °C
Mull lavas			
Zone G	8	0·19 ± 0·06	400–500 °C
I	17	0·32 ± 0·06	500–600 °C

* S_{200} refers to the stability of the remanent magnetization after a.f. demagnetization to a peak field of 200 Oe. The uncertainty is the R.M.S. error of the mean.

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Finally, the Curie point evolution diagram (Fig. 9) shows that regional hydrothermal alteration induced changes in thermomagnetic curves are quite differen from those resulting from deuteric oxidation. The former are characterized by single (conventional) Curie point curves and the latter, in the intermediate stages, by double Curie point curves.

Hence we conclude that the single high Curie point material represents *in situ* alteration of the original near-stoichiometric titanomagnetite without the formation of extra phases.

The problem then is to identify the structure and composition of this *in situ* altered titanomagnetite.

Microscopic examination shows the original titanomagnetite to be apparently unchanged until general alteration to a titanohematite begins to take place after burial to H or I zone conditions. Even when granulation is seen the remaining magnetite is usually an ordinary light brown colour. There is *no sign of a general colour change*, with increasing alteration, to the pale brown or off white cubic phase found irregularly replacing basaltic magnetite and usually known as maghemite or titanomaghemite. This is an important point as one of the mineralogical candidates for the replacement of titanomagnetite in the conditions of regional hydrothermal alteration is a cation-deficient spinel (CDS), of which a titanomaghemite seems the most likely to form.

In judging the experimental evidence for the altered titanomagnetites of basalts being in the CDS state it is necessary to try to resolved apparently conflicting reports of CDS properties.

Ozima & Larson (1970) describe the thermally induced breakdown of titanomagnetite to titanohematite and to essentially non-magnetic hematite and rutile. Inversion to these phases contrasts with the thermally induced changes in naturally altered titanomagnetites. Heat treatment of naturally altered titanomagnetites often leads to much greater specific moments and does not lead to a loss of more than half the original moment even in the case of the kink characterized H and I zone material.

However, Readman & O'Reilly (1970) have produced CDS material from a range of compositions on the magnetite–ulvospinel join which in many of their properties do match the properties of the single high Curie point magnetic phase of regionally hydrothermally altered basalts. Thus, Curie points rise with production of the CDS state. Inversion produces a range of phases, including near pure magnetite, and is accompanied by strong field heating curve changes similar to those found for natural material. Again the estimated time constant for the natural production of CDS material of 10^6 yr at 300 °C approaches estimates based on geological information.

In contrast, Sanver & O'Reilly (1970), in considering the implication of producing a non-stoichiometric, anomolously high Curie point magnetic phase by the laboratory heating of basalts, conclude that the natural conditions required to produce these changes may not occur. However, this conclusion may be based on a lack of knowledge of the temperatures and time intervals involved in regional hydrothermal alteration and thus might well be modified. While no final identification of the naturally occurring altered titanomagnetites with CDS phases can yet be made it appears likely that this identification will eventually prove to be correct. Clearly there is need for further experimental work as no inversion processes have yet been described in which CDS phases are partly replaced by ferri-rutile granules or sphene, or are wholly replaced by titanohematite, all of these situations being common in naturally altered basalts.

5.1.2 *The magnetic phases responsible for the complex thermomagnetic curves found for many H or I zone basalts.* No single phase is known to have the complex thermomagnetic properties often exhibited by material from these high alteration zones.

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This is not surprising since general alteration of basalts takes place in H/I zone conditions. Not only are the opaques altered into several new phases as described in Section 3 but in addition near pure magnetite or hematite is produced by the breakdown of any remaining olivine. Moreover, the secondary minerals epidote, prehnite, pumpellyite and ' chlorites ' are produced and some of these could break down to produce iron oxides or hydroxides during laboratory heating.

At the present time no candidate can be put forward to explain the kink feature of thermomagnetic curves for material in these high states of alteration. However, the ' e ' type Curie points and the decrease in moment with monotonous thermomagnetic curve, on cooling (Fig. 9, Section 4.2) are consistent with the initial presence of a nearly pure maghemite (Readman & O'Reilly 1970). These authors show that the inversion of wet ground oxidized pure magnetite shows both of these features with the inversion marked by a typical ' e ' type Curie point between 350 and 400 °C as is often found for H/I zone basalts.

The question remains as to the source of this nearly pure maghemite. It seems possible that it could be formed either from the products of the previous inversion of a high titanium CDS, one of which is found by Readman & O'Reilly to be an iron-rich spinel (presumably close to pure magnetite) or by the formation of a low titanium CDS from the by-products of olivine decomposition.

Evidence from H/I zone basalts suggests that the explanation involving the inversion of a low titanium CDS originally derived, via a high titanium CDS, from a high titanium titanomagnetite seems likely to be correct. Thus, both the Mull I zone lavas, in which magnetite (or a CDS derivative) was produced by olivine breakdown, and the Keweenawan I zone lavas, in which hematite was produced by olivine breakdown, show the complex thermomagnetic behaviour associated above with the presence of a near pure maghemite. Both sets of lavas often contain altered titanomagnetites in which an apparently isotropic brown phase is speckled with anisotropic white flecks probably consisting of a titanohematite. It is possible that this situation represents inversion of a high titanium CDS to a low titanium spinel plus a rhombohedral phase, with later CDS alteration of the spinel.

If the identification of a near pure maghemite in H/I zone basalts proves to be correct, as it seems likely to do so, the range of conditions in which CDS phases can be produced in basalts will be considerably extended. Study of the whole range of CDS production conditions and the range of CDS phases formed is clearly a major necessity if the natural magnetization of rocks is to be clearly understood.

5.2 *The effect of regional hydrothermal alteration on the original thermoremanent magnetism of basalts*

From the point of view of the usefulness of palaeomagnetism as a geologic tool consideration of possible changes in remanence direction must have priority. There is theoretical evidence that large directional changes are possible, thus, O'Reilly & Banerjee (1966) show that rather extreme oxidation of a high titanium titanomagnetite without phase splitting could result in a self-reversal of remanent moment.

Two lines of attack can be made on this problem. One uses laboratory thermally-induced changes in natural remanences and the other uses internal tests of palaeomagnetic data. Laboratory experiments of several types are relevant here. For example Ozima & Ozima (1967) show that by careful heat treatment, the net remanence acquired by submarine lava specimens can be made to oppose the magnetizing field. These authors find that heating to within the temperature interval 300–330 °C for less than 150 min defines the range of conditions for which self-reversal is produced. We must ask whether in natural conditions a reheating temperature of say, 200 °C, which must have been experienced by many lavas, for a very much longer period of time, might not produce self-reversal of remanence in some lavas. Creer & Petersen (1969) have produced partial self-reversal in low deuteric oxidation mag-

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netites by laboratory heating to 285 °C for periods from 4 to 462 min. After heat treatment the magnetites are still apparently optically homogeneous but strong field heating curves show double Curie points. Thus, the laboratory alteration is different from that produced in natural conditions. Creer & Petersen reconcile their optical and magnetic results by assuming that submicroscopic exsolution has in fact taken place. Similar results are reported in Creer, Petersen & Petheridge (1970). Again, Wilson & Smith (1968) and Marshall & Cox (1970) have studied changes in remanence direction during laboratory heating induced Curie point rises in basalts. In both cases new remanence, resulting from Curie point rise, is closely tied in direction to the original remanence. No evidence of self-reversal is reported. This is a surprising and encouraging result but its relevance to natural heating is somewhat in doubt as the peak temperatures used in laboratory heating are usually appreciably higher than those most commonly experienced during natural burial.

Turning now to palaeomagnetic results to test for the possible existence of regional hydrothermal alteration induced remanence direction changes. The classical tests for self reversal are useful here. So far as burial to Zone G conditions are concerned the evidence suggests that original TRM directions *can* be extracted from NRMs by suitable cleaning. Thus, lava-baked rock agreement, and within-lava directional agreement for a range of deuteric oxidation states are characteristic for zones A to G. The use of these classical tests is justified here on the grounds that high deuteric oxidation basalts (and possibly red sediments) are little altered by burial to Zone G conditions. However, it is difficult to make such a good case for lavas that have experienced Zone H or Zone I burial conditions. The problems here are that only a few well-documented studies of lavas in this H/I zone state are available and that high deuteric oxidation basalt, which appears to be little changed magnetically by burial to Zone G conditions, now also shows signs of alteration. Evidence from two lava areas does lend support to the possibility that original TRM information *can* be obtained from these extremely altered lavas. Palmer (1970) finds that the Zone I Keweenawan lavas can be subdivided into a basal ‘ reverse ’ group, found at least at three localities, and an higher ‘ normal ’ group. Evidence is adduced that a hard secondary component of magnetization is superimposed on the original magnetization with a secondary to original ratio of about 1 to 2. However, it seems to be possible to isolate the original direction by the correct analysis of the data. Again, Hubbard (private communication), in a detailed study of the Keweenawan North Shore volcanics from Grand Portage, Minn., finds that individual specimens from each flow group tightly, yet successive flows have distinct mean directions. This type of behaviour of flow mean direction of magnetization is well known for young little-buried lava sequences. Finally, the direction for both the Zone G and Zone I lavas of Mull cover a similar range, all being reversely magnetized with a tendency to directions shallower than the present centred axial dipole field for the area.

Summarizing, evidence for remanence directional change as a result of regional hydrothermal alteration is limited, but the possibility of its occurrence in special conditions cannot be ruled out until more evidence is available.

5.3 *Chemical effects of regional hydrothermal alteration relevant to the potassium–argon dating of basalts*

There appear to be two main ways in which the regional hydrothermal alteration of lavas could be a nuisance from the potassium–argon dating viewpoint. The first concerns the effect of reheating alone. Does the heating of a buried lava from 100 to 300 °C for prolonged periods of time several million years after eruption result in significant argon loss? The second concerns the redistribution of elements that takes place during the alteration process. The material of the zeolites and other secondary minerals is held to originate in the pyrogenic minerals of the lava itself.

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It seems likely that the source of the calcium, aluminium and silica of the zeolites is in the felspars or in the glass phase of basalts, and these may also be the sites of the small amounts of potassium presented. If this is so it is likely that in some cases at least the measured age of a basalt may in fact be the age of secondary mineral formation.

Laboratory experimentation suggests that temperatures in the zeolite facies range are unlikely to produce significant argon loss (Reynolds 1957; Gerling, Levsku & Morozava 1963). However, extrapolation from these results to geological time intervals may be unjustified at least in some instances.

Some natural experiments may also be relevant here. Thus Westcott (1966) has studied the argon loss from a schist as a result of reheating by dyke intrusion. Significant argon loss occurs, as was also found by Hart (1964) in an analogous investigation. However, the temperatures involved in both these natural experiments were, for a large part, much higher than occur in regional hydrothermal alteration, and chemical conditions (rock composition, water availability) were also quite different from those existing in a basalt lava pile. Westcott does make what could be a relevant point; that laboratory determined apparent activation energies are usually higher than those determined from natural (geological) experiments. Baksi (1970) reports an experiment which is relevant to the uncertainties concerning the chemical changes in a lava during secondary mineral formation. An apparently satisfactory K/A age was obtained for a Neogene basalt containing calcite-filled vesicles, the calcite having been removed by hand-picking following crushing of the rock.

The state of the glass in basalts if often taken as a criterion of the suitability of the rock for dating (Doell, Dalrymple & Cox 1966; McDougal, Allsop & Chamalaun 1966). While it is generally known that basaltic glass is readily altered by post-eruptive processes no study directly relating glass alteration to regional hydrothermal alteration is known to the authors. However, the work of Hay (1963) gives a clear picture of the effect that zeolite formation can have on volcanic glass. The 1000 m thick John Day formation of Mitchell, Oregon, consists of late Oligocene to early Miocene massive tuffaceous claystones, vitric tuffs and rhyolitic ignimbrites. The formation is sharply divided into two zones, an upper zone from which zeolites are absent and where fresh glass occurs and a lower zone in which the zeolite clinoptilolite replaces vitric (glassy) material. Zeolitization is dated as being subsequent to period of folding and prior to the eruption of the Mid-Miocene Columbia river basalts.

The results of an investigation of the variation of apparent age with specimen alteration in the Permo-Carboniferous doleritic Whin Sill of northern England (Miller & Mussett 1963) is also relevant here. In this case the apparent age was inversely related to groundmass alteration. The groundmass, where most of the potassium was located, consisted of a fine matrix of iron oxides and potash felspar forming a mesographic intergrowth with quartz. In extrusive basalts of the same composition as this sill the potassium-bearing equivalent of the groundmass would be glass (Dalrymple & Lamphere 1969).

From these two investigations it is clear that volcanic glass may be replaced by secondary minerals during regional hydrothermal alteration and that apparent age may depend strongly on the degree of groundmass glass alteration.

There is another manner in which regional hydrothermal alteration may result in young ages for basalts. Reliable use of the fission track dating method requires that a fraction of the fission tracks have not been lost by annealing. It seems likely that the higher temperatures reached during regional hydrothermal alteration could produce a degree of annealing (Aumento 1969), and this possibly deserves investigation.

5.4 *Conclusions*

It is impossible to answer completely any of the three questions posed in this section. The most complete answer can be given to the question concerning the

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reliability of palaeomagnetic data. The conclusion here is an encouraging one.

So far as the changes to magnetic minerals, and the production of new minerals, as a result of regional hydrothermal alteration, are concerned the need is partly for new techniques. Reflection microscopy, with magnifications of the order of × 1000, has been an ideal tool to elucidate the mineralogical changes caused by deuteric oxidation. Identification of the changes in titanomagnetite caused by the regional hydrothermal environment will probably need other techniques.

The biggest uncertainty concerning a possible deleterious effect of hydrothermal alteration involves its unknown influence in potassium–argon dating. The evidence is so limited that no estimate of the nuisance value of the alteration process can be made and experimentation is urgently needed here.

6. Summary and conclusions

In this paper we have described in some detail the opaque petrological and magnetic changes produced in basalts by a process which we propose to call regional hydrothermal alteration. The environment in which this alteration process acts is that of the zeolite metamorphic facies, for which maximum temperatures and pressures are about 300 °C and 1 kb respectively. While *local* hydrothermal activity is well known the point we wish to emphasize is the *regional* nature of the action of this alteration process. Thus some Recent and Quaternary, most Tertiary and all Pre-Tertiary basaltic rocks may be expected to have been affected by the alteration process.

As a result of our special interest in palaeomagnetism and the magnetic properties of basalts we have concentrated our attentions on the changes in the opaque minerals produced during burial by younger flows and saturation by warm ground water which constitute the zeolite facies environment. However, it is to be expected that the silicates also suffer changes; the growth of zeolites and other secondary minerals, presumably using material from the pyrogenic minerals, bears this out. A wide range of opaque mineralogical changes has been described. Among these are the formation of patches of fine ferri-rutile granules (granulation) in moderately altered high titanium titanomagnetites, and the alteration of titanomagnetite to titanohematite and the replacement of ilmenite by sphene in more altered basalts.

Changes in strong field thermomagnetic properties also follow the increasing degree of alteration. At first change is limited to gradual Curie point rise in basalts that have missed deuteric oxidation during initial cooling. With more extreme alteration the strong field curves for all basalts in all states of deuteric oxidation show evidence of changes in the magnetic constituents.

The recognition of a previously undescribed alteration process in basalts requires that the validity of conclusions based on palaeomagnetic and radiometric dating measurements be reassessed. This we have attempted to do with the following conclusions.

(1) Natural remanent magnetization can, by the application of suitable cleaning methods, be made to yield original cooling thermoremanence directions. This is a surprising but satisfying conclusion in view of the effort put into palaeomagnetic work over the last 20 years.

(2) The effect on radiometric ages of the chemical changes and physical history of basalts which have experienced the regional hydrothermal alteration process **is** little known and urgently needs study.

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