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SOURCE OF STABLE REMANENT MAGNETISM IN LAMBERTVILLE DIABASE

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ABSTRACT. Measurement of natural, thermal, and saturation remanent magnetism and their stability with respect to alternating field and thermal demagnetization of concentrates of plagioclase, pyroxene, and oxide (80 percent ilmenite, 20 percent magnetite) separated from a sample of diabase from the Triassic Lambertville (N.J.) sill, reveal that the stable remanence of the whole rock is associated with the silicates and especially with the plagioclase. Measurement of Curie-point and saturation moment of the feldspar indicates the presence of about 0.3 percent Fe ion in solid solution and 0.004 percent pure magnetite as a discrete phase. This magnetite cannot be identified microscopically, nor with an electron probe, and it is inferred to be present as submicroscopic, single domain particles.

INTRODUCTION

The presence and composition of ferrimagnetic oxides in rocks can usually be determined by either (1), direct microscopic observation (with the possibility of microprobe analysis), or (2), measurement of magnetic properties on whole rock or magnetic concentrates (Lindsley, Andreasen, and Balsley, 1966). These latter measurements (see Collinson and Girdler, 1967) fall into two categories: (2A)—those made in high magnetic fields, such as magnetic moment as a function of applied field (J/H), or saturation moment as a function of temperature (Js/T) which gives Curie points (Tc); and (2B)—those involving measurement of remanent magnetism (R.M.) (with alternating field = A.F. and thermal demagnetization), or low-field susceptibility, (X), usually performed and measured in field-free space, or simply in the Earth's field.

In high-field measurements on igneous rocks, properties compatible with those of a titaniferous magnetite (Tc $<580°C$, high X, low H_{sat}) with or without a second magnetic phase are almost invariably obtained. (Note: While members of the ilmenite-hematite series may have similar Curie temperatures, they have lower susceptibility, and a very high field is needed to effect saturation (H_{sat}).) As titaniferous magnetite is a common accessory in igneous rocks (Newhouse, 1936) and is almost invariably present in lavas (Carmichael and Nicholls, 1967), consistency between the results obtained by observation (1) and high-field magnetic measurements (2A) is usually found. Sometimes, however, these are inconsistent, as in the disagreement found in some basalts by Ade-Hall (1964) and Smith (1967) between the measured Curie-temperatures and those predicted from microprobe determination of the composition of optically homogeneous accessory spinels. Smith attributed this discrepancy to a subtle metastable oxidation.

The methods involving measurement of remanent magnetism (2B) are not directly diagnostic of the ferrimagnetic phase, unless perhaps

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thermal demagnetization is performed. This however, is not standard practice with igneous rocks, because of the oxidation to which spinels are prone when heated in air. Thus, it has been customary to *assume* that the remanence of the rock is carried by the ferrimagnetic phases seen and implied from high-field measurement (1 and 2A).

It is the larger accessory oxide grains ($>10\mu$) that are most readily noticed, studied, and analyzed in polished surfaces (1). As the bulk of the ferrimagnetic-phase in the rock is contained in these large grains, they will undoubtedly dictate the bulk magnetic properties (measurements 2A). The question remains whether these grains are indeed the carrier of the N.R.M. or more specifically the stable component of N.R.M.

The interest of the senior author in this problem was aroused first by examination of polished sections of specimens of norite from the Great Dyke in Rhodesia, used by McElhinny and Gough in their paleomagnetic study (1963). The only accessory oxide to be seen in some specimens which had N.R.M. of normal intensity ($>10^{-4}$ emu/cc) was one phase ilmenite (paramagnetic at room temperature). Subsequently, a similar anomalous situation was observed in paleomagnetically stable dolerite samples from the Roraima district in South America (Hargraves, 1968). In these dolerites, in fact, samples with significant accessory magnetite were characterized by higher bulk susceptibility and instability (p. 153). Where then was the ferrimagnetic phase responsible for the remanence, and why was it so stable?

A simple procedure was developed, designed to allow investigation of the natural remanence of the individual minerals in rocks, except insofar as their magnetic properties are modified by the crushing process needed to separate them and sedimenting needed to reorient them. The technique was employed by Young (ms) in a study of some red beds and a diabase sample, and a preliminary account of the results was presented at the I.A.G.A. Meeting in 1967 (Young and Hargraves, 1967).

This paper presents the results obtained using these techniques from a more intensive investigation of the natural remanence of minerals in a sample of diabase from the Lambertville sill (N.J.). N.R.M. measurements were supplemented by investigation of T.R.M. and saturation R.M. and their stability with respect to alternating field (A.F.) demagnetization.

PETROGRAPHY

The Lambertville Sill (1800 ft thick) is one of a number of diabase intrusions and extrusions associated with the Triassic Newark series of New Jersey (for example, Palisades, Rocky Hill Sills, and the Watchung Lavas). The sample studied was collected in a quarry at a point 750 to 800 ft up from the base in the exposures paralleling the Delaware River. It consists essentially of plagioclase (An 50) 61.0 wt percent, augite 35.1 wt percent, and oxide 3.9 wt percent, comprising coarsely intergrown titaniferous magnetite and ilmenite together with discrete ilmenite (approx 80 percent ilmenite, 20 percent magnetite).

EXPERIMENTAL PROCEDURE

In general, samples are crushed in a porcelain mortar, washed, screened, and in an appropriate size range, the mineral phases separated using heavy liquids. The silicate concentrates may be further purified if necessary, by treatment with hydrochloric acid, which will remove any contaminating magnetite exposed at the surface. The mineral particles are then reoriented according to their natural remanence (if any) by settling in a magnetic field in a solenoid 120 cm long imparting a "detrital" N.R.M. (D.N.R.M.).[1] The sedimentary media utilized ranged from melted paraffin wax, water-soluble Carbowax, and silica gel (Ludox). Freezing of the wax or gelation of the Ludox provides samples in a form suitable for measurement of the remanence and coercivity spectra. While the mineral powder could easily be recovered from the Carbowax and used for other tests if necessary, silica-gel was the only sedimenting medium suitable for thermal demagnetization. After dehydration, these samples are rather fragile and should be measured with an astatic or slow-speed spinner magnetometer. However, they may be heated to 710°C without harm.

Our attempts to investigate the natural remanence of the separated minerals were supplemented by measurements of intensity and stability of saturation remanence (8000 gauss) and the T.R.M. induced by cooling from 600° in a 1.04 oersted field. For these latter experiments, the sample preparation technique of Evans, McElhinny, and Gifford (1968) was used, whereby mineral powders are mixed with KBr and formed into cylindrical capsules by pelletizing the mixture in a mould under 1 kb pressure.

In the sample studied, the plagioclase, pyroxene, and oxide mineral fractions were separated from the −120 +200 mesh (74-125 μ) size range using Bromoform (S.G. 2.8) and Clerici solution (S.G. 4.0). The two silicate fractions were split, and alicquots treated with concentrated HCl for 9 hours. This resulted in small loss of weight and a distinct "whitening" of the plagioclase fraction. It is estimated that the concentrates are at least 99.0 percent pure. For some experiments, portions of the acid treated plagioclase concentrate were passed through a Frantz magnetic separator at maximum current (1.85 amps). This treatment removed a small fraction of composite plagioclase + pyroxene grains.

RESULTS

The intensity and stability with respect to A.F. demagnetization of the detrital N.R.M. of the three mineral phases and an uncrushed sample of the original rock is compared with their saturation remanence

[1] In previous experiments on powdered red-beds using this apparatus, fields of 30 gauss appeared to be sufficient to impart maximum D.N.R.M. intensity, and this field strength was routinely used in these experiments.

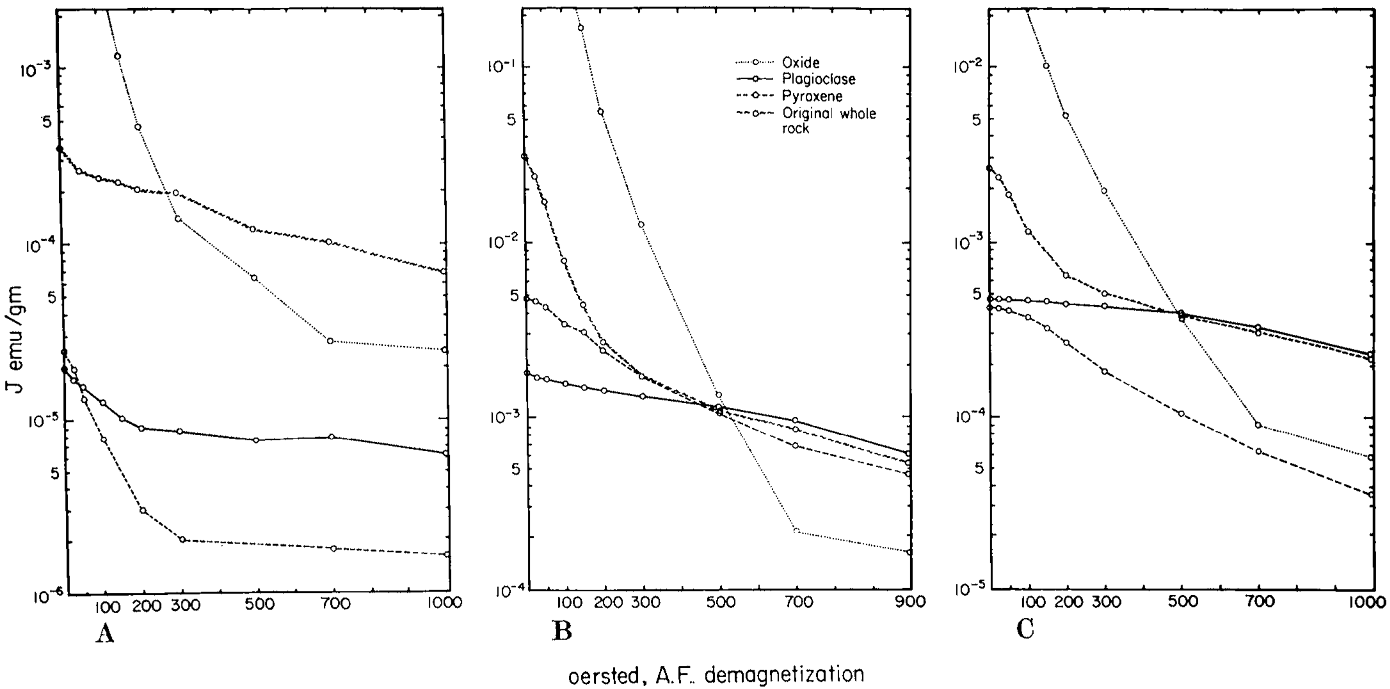


Fig. 1. Stability with respect to A.F. demagnetization of specific remanence intensities of pyroxene, plagioclase, and oxide mineral fractions, and comparison with uncrushed whole-rock. A. "Detrital" natural remanence—D.N.R.M.; B. saturation R.M.; C. thermal R.M. acquired on cooling from 600°C in 1.04 oersted field.

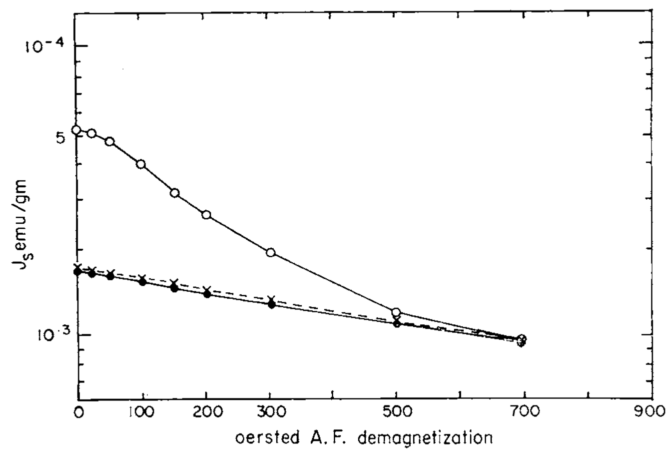


Fig. 2. Comparison of stability with respect to A.F. demagnetization of specific remanent intensity of plagioclase mineral concentrate. A. prior to acid treatment (solid-line, open circles); B. after treatment for 9 hours with concentrated HCl (solid-line, solid circles); and C. after further purification with Frantz magnetic separator (broken line). The slight, but anomalous increase of C over B is attributed to experimental error; the two are not significantly different.

and T.R.M., in figure 1A, B, and C. The saturation remanence of original and acid treated plagioclase concentrates is compared in figure 2 to illustrate the effect of such treatment. The intensity of the reconstructed rock (contribution of the separated minerals weighted according to their modal abundance) is compared with that of original rock in figures 3A, B, C. Finally, the percentage contributed to the total intensity of the reconstructed rock by the individual phases, as a function of alternating field demagnetization is shown in figure 4A, B, C.

DISCUSSION

In all experiments (D.N.R.M., T.R.M., and saturation remanence) it is clear that the remanent magnetism residing in the plagioclase is most stable, that in the pyroxene is intermediate, and that in the oxide is extremely unstable. Although the initial remanent intensity of the silicates is small compared with that of the oxides, their modal abundance and stability is such that above about 200 oersted, they contribute the bulk of the remanent magnetism. The contribution of the plagioclase alone (61.0 wt percent) is predominant above 200 oersted to 400 oersted and varies between 70 and 90 percent of the total above 700 oersted. It should be noted (fig. 1B, C) that above 500 oersted A.F., the *specific* saturation and thermal remanence of the plagioclase exceeds that of the oxide!!

FERRIMAGNETIC MINERALOGY

Oxides.—Ilmenite is the predominant accessory oxide phase, occurring as independent grains, and in part as coarse intergrowths with mag-

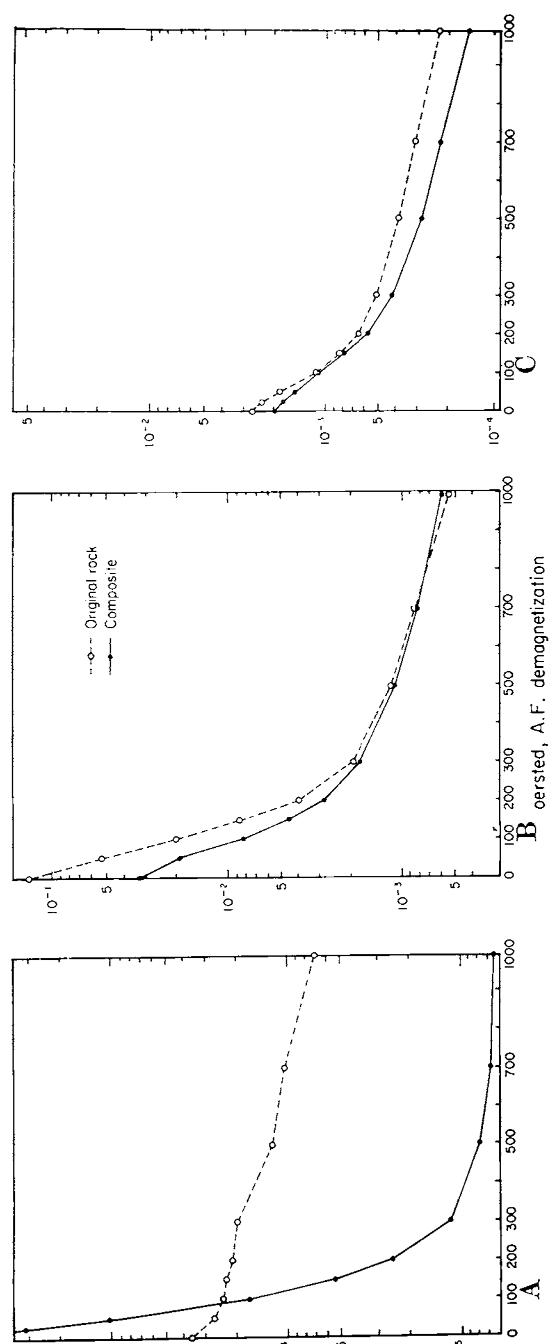


Fig. 3. Comparison of specific remanence intensity of uncrushed whole-rock (broken-line) with reconstructed rock (solid line) obtained by weighting specific intensities of mineral fractions according to their modal abundance. A. D.N.R.M.; B. saturation R.M.; C. TRM. Note agreement in (B) and (C) and conspicuous difference in (A).

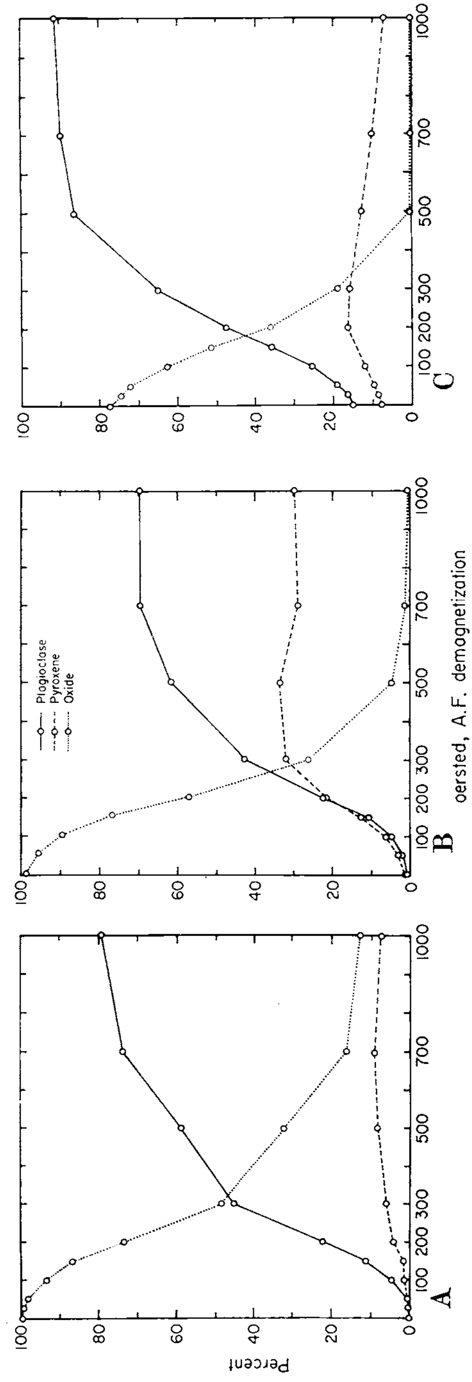
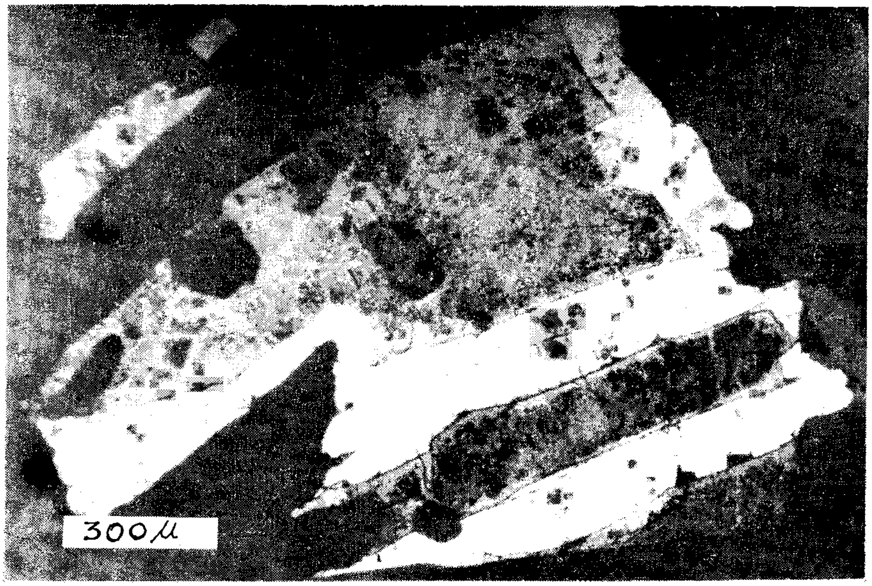
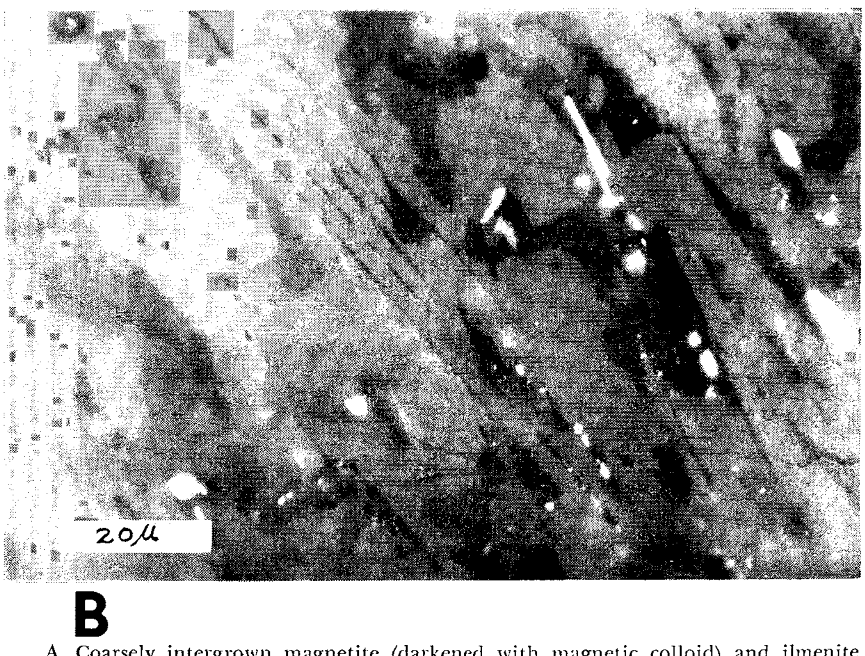


Fig. 4. Percentage contribution of individual phases to specific intensity of reconstructed whole-rock as a function of A.F. demagnetization. Plagioclase, solid line; pyroxene, broken-line; oxide, dotted line.

PLATE 1

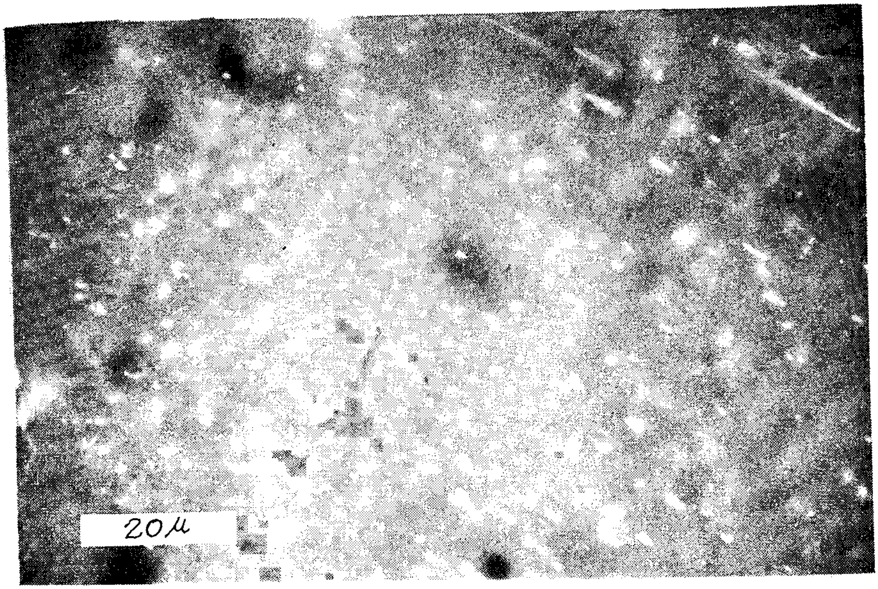


A

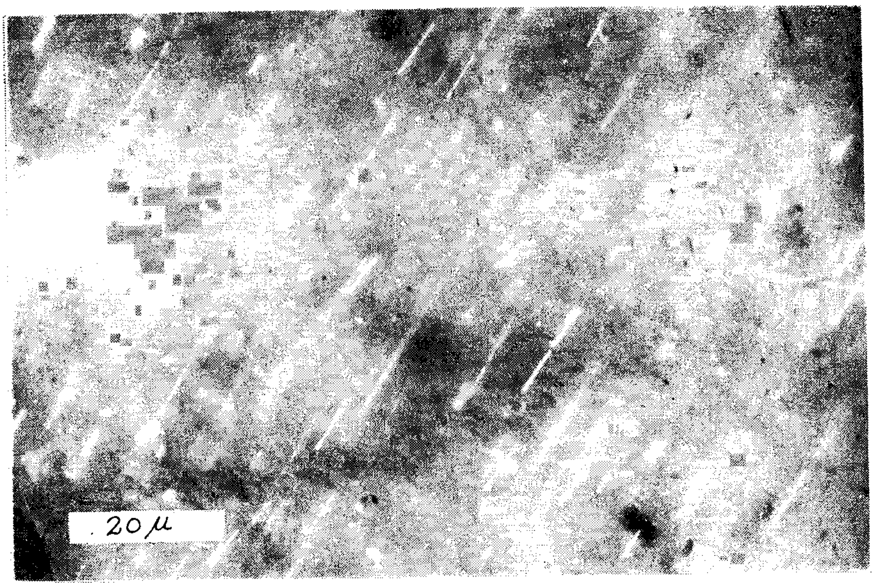


B

A. Coarsely intergrown magnetite (darkened with magnetic colloid) and ilmenite (clear) grain, B. Small magnetite particles in augite.



C



D

C. and D. Enigmatic non-opaque rodlets in plagioclase (see text).

netite (pl. 1-A), the grains vary between 0.2 and 0.5 mm in diameter (median grain diameter—m.g.d. = 0.26 mm). The Curie temperature of the magnetite (fig. 5) is 545°C (± 5°), suggesting the presence of a small amount of ulvo-spinel in solid solution. The saturation magnetization of the oxide concentrate is about 14 emu/gm, suggesting the presence of about 20 percent magnetite (only 15 percent if it were pure Fe_3O_4). Thermal demagnetization of T.R.M. (fig. 7) shows a predominant blocking temperature just below 550°C, compatible with the Curie point measurement.

Pyroxene.—The augite (m.g.d. = 0.46 mm) contains distinct small opaque particles (pl. 1-B) up to several microns in diameter. Magnetic colloid adheres to these, suggesting that they are magnetite. The Js/T curve (fig. 5) is dominated by the paramagnetic Fe in the silicate structure, but a slight inflection does occur between 500° and 600°, indicating the presence of a ferrimagnetic phase. The J/H curve (fig. 6) is likewise dominated by the paramagnetic susceptibility, but the ferrimagnetic phase may have saturation moment of 2 x 10^{-2}emu/gm, equivalent to 0.02 percent magnetite. Thermal demagnetization of T.R.M. in the pyroxene (fig. 7) indicates a phase with a blocking temperature close to 575°, the Curie point of magnetite.

Plagioclase.—The plagioclase (m.g.d. = 0.34 mm) is largely fresh and translucent although some grains contain brownish turgid alteration patches. The only unusual "particles" noted in thin and polished section of whole-rock and powder were extremely small (<1 μ diam) rodlets visible in some orientations of completely fresh feldspar grains (pl. 1-C, D). Magnetic colloid, however, did not appear to adhere to these rodlets.

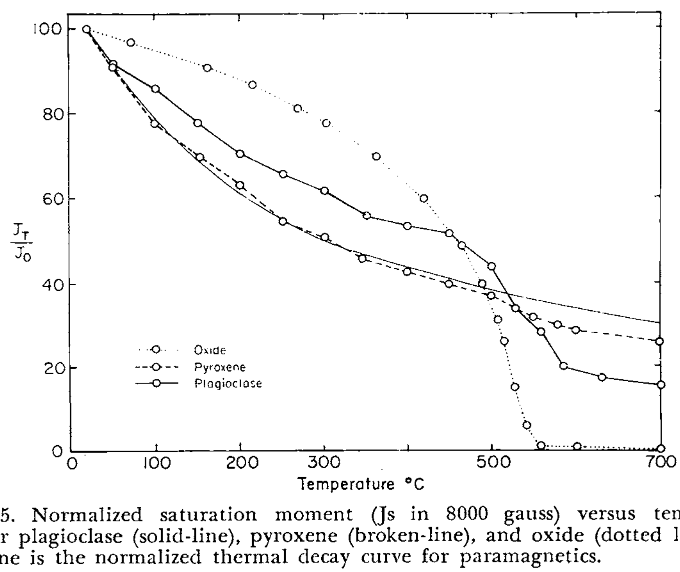


Fig. 5. Normalized saturation moment (Js in 8000 gauss) versus temperature curves for plagioclase (solid-line), pyroxene (broken-line), and oxide (dotted line). The curved-line is the normalized thermal decay curve for paramagnetics.

As they could be seen to extend *obliquely* below the surface of the polished section (pl. 1-C, D), they cannot be opaque and still reflect light *vertically*. Thus the illumination must result from internal reflections in a transparent phase. Areas of feldspar containing these rodlets were scanned for Fe, K, Ti, Si with an electron probe (A.R.L., Model Emx-sm, beam diam ¼ μ) and showed no concentrations suggesting the presence of a separate phase containing these elements. The precise nature of these rodlets remains unknown.

The Js/T curve of the plagioclase (fig. 5) indicates the presence of a ferrimagnetic phase with Curie temperature between 550° and 600°C (approx 575°) suggesting pure magnetite. This fact clearly distinguishes the ferrimagnetic phase in the feldspar from the primary accessory magnetite (Tc $<$550°) proving that the ferrimagnetism of the feldspar is not due to contamination from primary oxide. The initial remanence of the plagioclase is comparable to that of the pyroxene (and considerably more stable); thus contamination of the plagioclase by pyroxene cannot be the cause of the former's ferrimagnetism (compare Evans, McElhinny, and Gifford, 1968, page 144).

The J/H curve of the plagioclase (fig. 6) suggests the presence of two magnetic components: one ferrimagnetic, saturated in about 2500 oersted, and a second paramagnetic component—indicated by the progressive increase in moment in fields above 2500 oersted.

The Js of the ferrimagnetic phase is about 3 x 10^{-3}emu/gm. If this is indeed pure magnetite (Js $\cong$ 100 emu/gm) it indicates an abundance of about 0.003 percent!!

The paramagnetic susceptibility of the second component is about 6 x 10^{-7} emu/gm oersted. This is equivalent to an Fe^{3+} content of about

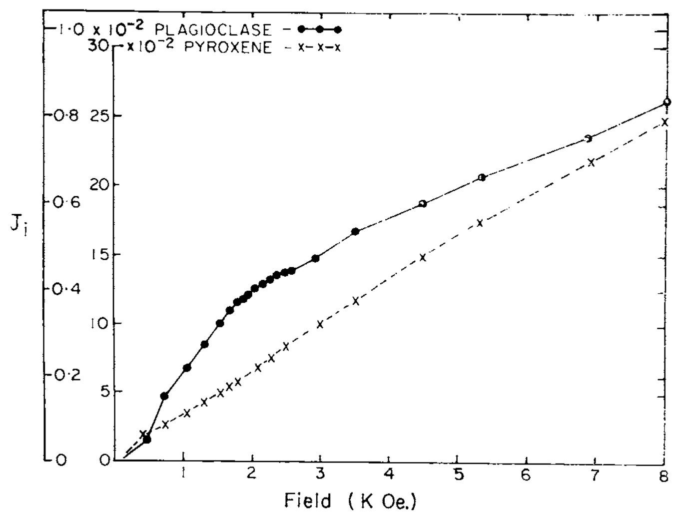


Fig. 6, Specific magnetic moment versus applied field curves for plagioclase (solid-line) and pyroxene (broken-line). Note contrasting Ji scales for the two minerals.

0.3 percent, expressed as Fe_2O_3 (Collinson, 1968). The feldspar has not been analyzed for iron, but 0.3 percent Fe_2O_3 is a normal value for intermediate plagioclases (Corlett and Ribbe, 1967). What is more noteworthy is how little of the total iron in the feldspar is apparently contained as magnetite.

As the magnetite cannot be seen microscopically, and its presence is only inferred indirectly, it is reasonable to speculate that it may be present as single domain particles. If the easy-directions of these particles are randomly oriented, the saturation remanence should be half the saturation moment (3-4 x 10^{-3}emu/gm). The measured initial saturation remanence (fig. 1B) is 1.75 x 10^{-3}emu/gm, in agreement with the predicted value.

Thermal demagnetization of T.R.M. in the plagioclase (fig. 7) shows that it is much more stable than either pyroxene or oxide, In view of the distinct difference in the Curie temperature of these two phases, however, (fig. 5), the relatively low Tb of the plagioclase cannot be due to contamination. As blocking temperatures are dependent on grain volume in single domain particles (Neél, 1949), the low Tb of the plagioclase may be judged to indicate the presence of ferrimagnetic particles, somewhat smaller than the maximum single-domain size. For pure magnetite, Neél

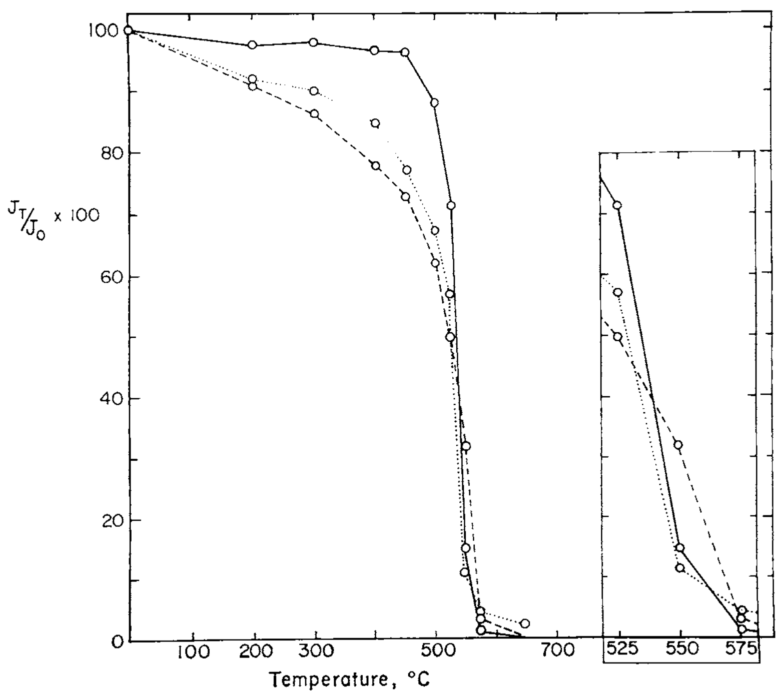


Fig. 7. Thermal demagnetisation of TRM (acquired by cooling from 600°C in 1.04 oersted field) of plagioclase (solid-line) pyroxene (broken-line) and oxide (dotted-line). Inset is enlargement of demagnetization curves between 500° and 600°C.

(1955) has calculated this size to be about 0.03 μ. If the particles are elongate this will increase (Nagata, 1961, p. 21; Strangway, Larsen, and Goldstein, 1968); in any case, however, single domain particles of pure magnetite are likely to be beyond the limit of resolution by optical means.

In this feldspar, the intensity of T.R.M. acquired by cooling from 600°C in a field of 1.04 gauss is 4.6 x 10^{-4}emu/gm (fig. 1C). This is roughly one quarter of the saturation remanence or one eighth of the total saturation moment! It remains to be determined whether the rule of linear relation between low-field strength and T.R.M. (Nagata, 1961, p. 153), on which all paleointensity studies depend, still applies in this material.

EFFECT OF CRUSHING ON THE MAGNETIC PROPERTIES OF THE MINERAL PHASES

Whereas the specific intensities of T.R.M. and saturation remanence of the "reconstructed" rock compare well with that of uncrushed material throughout the A.F. demagnetization spectrum, the intensity of sedimented "D.N.R.M." is quite different from the true N.R.M. (fig. 3). It had been our experience with red beds that a 30 gauss field (as used in this study) was sufficient to give maximum D.N.R.M. to the sedimentary powders. Therefore this discrepancy between D.N.R.M. of the powders and the N.R.M. must reflect either a change in magnetic properties of the minerals as a result of the crushing or the inefficiency of the experimental conditions as a means of reorienting the particles.

In this study the primary D.N.R.M. (to which the oxide fraction is the principal contributor) is greater than the N.R.M. (fig. 3); our qualitative explanation for this is as follows:

The particle size used in the experiment (−120, +200 mesh) averages about 100μ. This is considerably less than the m.g.d. of the accessory oxide grains (0.36 mm). Thus, the oxide grains with their original multi-domain configuration are broken down into smaller, but still multi-domain configuration fragments. The remanence of *all* these individual fragments are now aligned in the sedimentation process, the total being considerably greater than the vector sum in the original grain. Thus, if a large multi-domain grain is broken down into smaller multi- or single-domain particles, the maximum D.N.R.M. of the powder will be considerably greater than the original N.R.M.

The residual remanence after 500 oersted A.F. demagnetization, however, is considerably stronger in the natural rock than in the powder, and in this range, the silicates are the principal contributors.

Although the silicate grains used average 100μ the ferrimagnetic particles they contain are at least one order of magnitude smaller (presumed to be much less than 1μ in the plagioclase). The numbers of ferrimagnetic particles in the silicates that are broken during the crushing is negligible compared with those surviving intact within the silicate particles. The intensity of the R.M. of a unit volume of silicate particle should not be significantly changed by ferrimagnetic particle-breakage due to crushing. Thus either the silicates are demagnetized by the crushing process, or the particles are not well oriented.

To test the orienting efficiency of the experimental conditions used, fresh aliquots of the two silicates were sedimented in 80 gauss fields, the maximum available with our apparatus. After appropriate A.F. demagnetization, the D.N.R.M. of the pyroxene was found to be increased (relative to the original result, fig. 1A) by a factor of about 5, while the plagioclase was increased by a factor of only 1.4. Although the orientation had been improved by increasing the field, there remained a considerable discrepancy between the D.N.R.M. and uncrushed N.R.M. (compare fig. 3A). Furthermore why, on increasing the field, should the detrital moment of the pyroxene be increased more than the plagioclase?

Under standard experimental conditions, a prime factor influencing the efficiency of orientation will be the ratio of magnetic moment to the mass of the particle. This ratio (Jn. emu/gm/mass of 100μ particle) in the case of the oxide is more than two orders of magnitude greater than that in either pyroxene or plagioclase. On this basis one would expect the oxide to be more efficiently oriented than the silicates.

Another factor influencing particle orientation is their intrinsic shape anistotropy. As the pyroxene particles are significantly more equant than the somewhat tabular plagioclase crystals, the orientation of the former are more likely to be preserved (against gravity) on arrival at the bottom of the settling tube. This may explain the contrasting behavior of the two silicates on increasing the settling field-strength.

In terms of the data available, it remains uncertain whether to some extent the silicates, despite the minute size of their included ferrimagnetic particles, *have* been significantly demagnetized by crushing. The point is immaterial, however, with respect to the conclusions of this study. Nevertheless, further investigation of the problem is in progress, for if the magnetically stable component in a rock can be separated unharmed (magnetically), then development of an efficient reorienting technique would facilitate paleointensity studies.

SIGNIFICANCE OF RESULTS

The findings of this study are in keeping with the accumulating evidence that stable remanence in rocks resides in small, inconspicuous, possibly single-domain ferrimagnetic particles (Dunlop, 1968; Stacey, 1967).

Soffel (1968a,b) has investigated the efficiency of dislocations and inclusions in pinning domain walls and hence causing high coercive forces in multi-domain titaniferous magnetite grains (as is proposed in the T.R.M. theories of Verhoogen, 1959, and Ozima and Ozima, 1965). His conclusions as to the energy required for domain walls to surmount these obstacles, coupled with reasonable estimates as to their abundance in natural minerals, make it seem unlikely that this mechanism can account for much of the stable component of T.R.M. in rocks. Soffel (1968a,b) studied also the T.R.M. intensity and stability characteristics of two basalt samples which differed only in the average particle size of the homogenous titaniferous magnetite grains (Usp60 percent, Tc $\cong$ 200°C). The high intensity and stability of the fine grained sample relative to the coarse could be quantitatively explained in terms of the particle size spectrum, assuming only that grains less than 2μ were single-domain.

Strangway, Larsen, and Goldstein (1968) review the theoretical arguments associating the high stability components of N.R.M. with single-domain particles. In the case of basic volcanic rocks, they suggest that high temperature oxidation of titaniferous magnetite which produces ilmenite lamellae leads to the isolation of suitably shaped and sized magnetite particles in the host grain. They present experimental evidence demonstrating the increase in coercivity which results from heating lava specimens to 800°C in air. They associate this increase with the observed increase in ilmenite lamellae in the magnetite.

A striking demonstration of the relative unimportance with regard to remanence of the large accessory oxide grains is provided by the study of Watkins and Haggerty (1967) on the effect of oxidation on the magnetic properties of basalt. They found the most intense and stable remanence to occur in the most oxidized samples. In these, however, the primary titaniferous magnetite particles are completely altered to an intergrowth of rutile or pseudobrookite and hematite. The amount of hematite (the only "ferromagnetic" phase remaining) is far too small to explain the high remanence, which the authors attribute to small magnetite particles produced by oxidation in the olivine.

Evans and McElhinny (1966) and Evans, McElhinny, and Gifford (1968) in their study of the Modipe gabbro showed that the stable remanence was associated with small magnetite particles "exsolved on the cleavage planes of the pyroxene". The contribution from the plagioclase in this gabbro was relatively insignificant however and could be explained by a trace of pyroxene contamination in the feldspar concentrate.

Bergh (ms, 1968a, b) in his paleomagnetic study of the Stillwater Igneous Complex found that in rocks apparently devoid of a primary oxide precipitate, the anorthosites carried stable remanence, and that the susceptibility and magnitude of the secondary magnetic components in the N.R.M. increased with the pyroxene content.

These data, together with the results of this study, illustrate the diversity of possible sources of stable remanent magnetism. Most important, they demonstrate that stable R.M. may reside in submicroscopic particles in the silicates. Thus understanding of the magnetic properties of rocks, of especial importance in paleomagnetic studies, requires insight into the properties and petrogenetic history of the entire mineral assemblage, not simply the accessory oxide phases.

ACKNOWLEDGMENTS

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