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The Role of Pigment and Specularite in the Remanent Magnetism of Red Sandstones

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Summary

The origin of the remanent magnetism of red sandstones has been investigated by separating the polycrystalline haematite grains (specularite) from rocks and comparing the properties of the remanent magnetism of the specularite, the residual pigment-quartz separate and the original rock. From the results it is possible to determine the relative importance of the two haematite forms as carriers of the remanent magnetization in different rocks and to suggest probable magnetization processes.

1. Introduction

Although red sandstones have provided much reliable palaeomagnetic data, the process by which a particular formation acquires its natural remanent magnetization (NRM) is not generally known, nor the relative importance of the two forms of haematite, the red pigment and black, crystalline specularite, as carriers of the NRM. Previous work by the author (Collinson 1966a, 1968) established some features of the mineralogy and iron oxide content of red rocks in relation to their bulk magnetic properties (e.g. isothermal and induced magnetization, initial susceptibility) but any correlation between intensity of NRM and other magnetic properties or iron oxide content has rarely, if ever, been found. This is not unexpected since in any rock the intensity of NRM will clearly depend on several variables including the absolute and relative amount of pigment and specularite (both of which can carry remanence), their specific intensity of magnetization and the degree of alignment of the magnetization axes of the particles contributing to the NRM. Also, bulk magnetic properties and iron oxide content are due not only to haematite but also by other iron-containing minerals such as ilmenite ($FeTiO_3$) and clay minerals; there are paramagnetic and do not carry remanence.

Investigations of specific formations have hitherto provided some information. Using a chemical demagnetization technique Collinson (1965) showed that samples of the Chugwater formation (Triassic) from the western USA owed the bulk of its NRM to their specularite content, and that the Taiguati formation (Permo-Carboniferous) from Bolivia was magnetized at least partly by the pigment (Collinson 1966b). Roy & Park (1972), using a similar technique, showed that the magnetization of the Hopewell Group sediments (Carboniferous) from New Brunswick, Canada was contributed by both pigment and specularite. By studying individual particles of specularite extracted from the Bonito Canyon quartzite (Pre-Cambrian) from Arizona, Collinson (1972) deduced that they carried the NRM, the wide range of intensities observed being due to varying scatter of the magnetization axes of the particles in different samples.

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Neither of the techniques used above is universally applicable to sediments and another approach is desirable which can be applied to a wider range of rocks.

There appear to be two processes by which red sediments can acquire a stable primary magnetization. In depositional (or detrital) remanent magnetization (DRM) magnetized particles (pigment of specularite) are aligned in the Earth's magnetic field while settling in water and this alignment is preserved during deposition and subsequent drying out and consolidation of the rock, thus imparting an NRM to it. An alternative process is chemical remanent magnetization (CRM) which results during the formation of a magnetic mineral by chemical or physical change in a magnetic field. Examples of possible processes in redbeds are the *in situ* oxidation of magnetite to haematite, the formation of red pigment by precipitation from iron bearing solutions and the dehydration of hydrated iron oxides to form haematite. It is not always possible to determine the genesis of pigment and specularite in sediments, and the relative importance of the above two processes has hitherto been unknown. There are no obvious differences expected in rocks which have been magnetized by one process or the other.

If the origin of the NRM is the object of an investigation, it is clearly desirable to confine the studies to the properties of the NRM, and, if possible, the separate remanent magnetic properties of the pigment and specularite. Unfortunately, techniques that can be applied to the NRM of the whole rock are limited, and crushing followed by separation of the pigment and specularite is not only difficult but simultaneously destroys the rock magnetization. In this paper a compromise solution to the problem is described which has provided information on the part played by pigment and specularite in the NRM of redbeds and has enabled the magnetization process to be inferred.

2. Techniques

The principle of the technique is to crush a sample of the rock, separate out the specularite by means of a combination of heavy liquid separation and slow-speed centrifuging and then to 'reconstitute' the remanent magnetism of the specularite and pigment-quartz separates by allowing them to settle separately in a liquid column in a magnetic field; the resulting sediments then possess a DRM which, after drying out, can be studied for their remanent magnetic properties and compared with the properties of the NRM of the original rock.

For the separation process, a 10 – 15 g sample of rock is crushed to pass a 300 mesh sieve (53μ) and a 2-g portion placed in a cylindrical glass container 4 cm in diameter and 15 cm high tapering at the lower end to a ground glass exit tube; this is closed by a glass rod ground at one end to fit the exit tube and extending above the top of the cylinder. The cylinder is filled with bromoform to a depth of about 5 cm, agitated to disperse the rock powder and then placed in a slow-speed centrifuge (5 rev s^{-1}, approximately 15x gravity) and centrifuged for 15–20 min. The heavy minerals, including specularite, accumulate at the base of the cylinder and can be collected by lifting the central tube slightly and allowing the minerals and a small amount of bromoform to run out. The centrifuging is repeated until no further minerals collect, and the procedure repeated with the remainder of the rock powder. The specularite is recovered by evaporation and the pigment-quartz residue by filtering and drying-out.

With some rocks this procedure produces a clean separate of dark opaque minerals, much of which is haematite; total iron determinations (as Fe_2O_3) give between 15 and 40 per cent. It is not easy to determine how much specularite is left in the pigment-quartz separate, but the relative weight of the specularite and the appearance of the pigment residue indicates that a substantial proportion has been extracted. In other rocks the separation is less well defined, particularly in those with high pigment content, and the heavy fraction includes some pigment; a pigment-covered quartz grain

containing about 10 per cent by weight of haematite (or more) will sink in bromoform. Although neither fraction is pure specularite or pigment, the remanent magnetic properties of each will be dominated by one component, and they are referred to hereafter as 'pigment' and 'specularite'.

The specularite and pigment separates are formed into depositionally magnetized sediments by allowing them to settle in a horizontal magnetic field in a 5-cm column of acetone which terminates in a shallow silica dish. 1·0–g portions of pigment were used and 10-to-50-mg portions of specularite, depending on the amount available. The specularite was deposited by itself, as obtained from the separating process. The choice of magnetizing field is governed by the requirement that it does not impart a detectable isothermal magnetization to the samples while at the same time it is strong enough to produce the maximum possible alignment in the DRM process. Preliminary tests showed that 10 0e was a satisfactory value. After the sediments were dried out they were found to be sufficiently compacted, if handled carefully, for the appropriate experiments to be done on them. All measurements of NRM and DRM were done with an astatic magnetometer.

3. Results

3.1 *Thermal demagnetization experiments*

All the rocks that are described in this paper have stable directions of magnetization

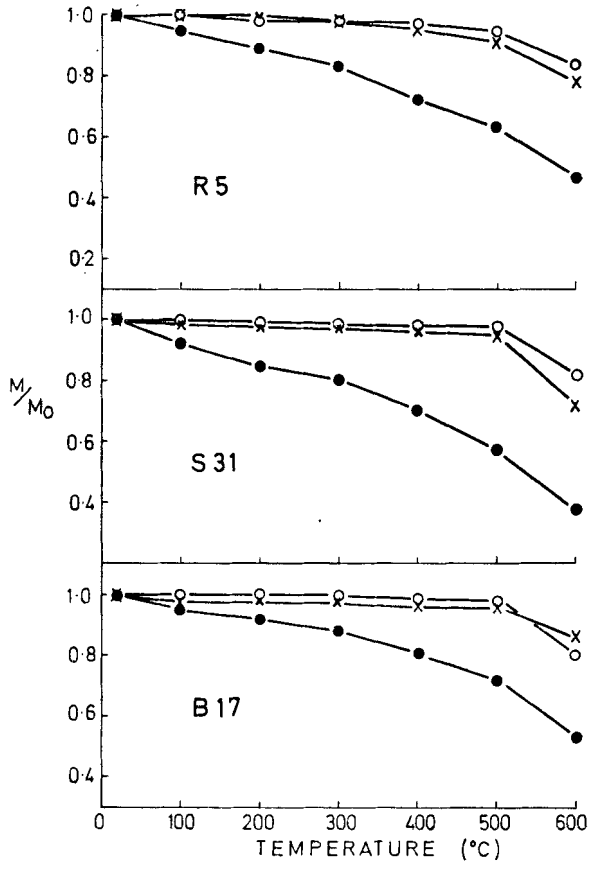

FIG. 1 Thermal demagnetization of the NRM of the rock and the DRM of extracted pigment and specularite from three samples.
rock, ○; pigment, ●; specularite, ×.

that show no significant change on heating. All the deposited samples of specularite and pigment possessed a measurable DRM, in the range 10–100 x 10^{-6} emu g^{-1} in the pigment and a much stronger 1000–6000 x 10^{-6} emu g^{-1} in the specularite. Preliminary tests were carried out on the alternating field and thermal demagnetization characteristics of the DRM; the former showed some differences between the pigment and specularite but there was a much more marked difference in the latter. With both techniques the DRM of the specularite was more stable than that of the pigment. It was therefore decided to compare the thermal demagnetization decay curves of the NRM of the original rock and the DRM of the pigment and specularite separates. The results fall into three groups, which are described below. All samples were heated in air in field-free space to the particular temperature required, allowed to cool and their magnetization measured.

Group I. These rocks comprise a series of fine sandstones and siltstones, usually pale orange-red-brown in colour (near 5 YR 5/6, 10 R 6/6 and 10 R 5/4 on the Munsell rock colour chart) with a low to moderate pigment content and specularite varying in amounts and grain-size. The separation process produces a well-defined dark coloured heavy fraction 1–3 per cent by weight of the original sample. The results of thermal demagnetization of the DRM of the separates and the NRM of the rock are shown in Figs 1 and 2. There is a marked difference in the hardness of the magnetization of the

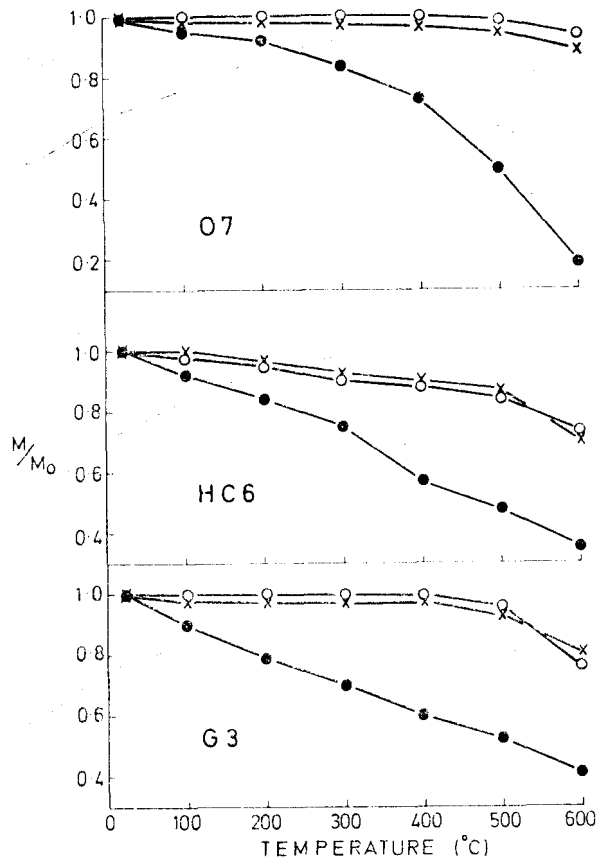

FIG. 2 Thermal demagnetization of the NRM of the rock and the DRM of extracted pigment and specularite from three samples. Symbols as in Fig. 1.

pigment and specularite fractions, with the latter always much more stable and the decay curve for the rocks follows that of the specularite rather closely.

Although there is evidence that the specularite is carrying the NRM in these rocks, it is desirable to investigate the results further. The efficiency of the separation technique in extracting the specularite is difficult to assess and there may be sufficient left in the pigment fraction to affect the shape of its decay curve; for this reason the true pigment decay curve might be expected to be more rapid than the ones shown. Another question is whether there is a magnetically hard component in the pigment which could carry the remanence in the rock but which is obscured in the DRM sample because a greater quantity of 'soft' material which is not aligned in the rock but is aligned by the DRM process. Alternatively, are the 'hard' magnetic particles which dominate the DRM of the specularite randomly directed in the rock, and are there some particles with a soft magnetization as well?

A possible way of detecting particles with differing magnetic properties in each fraction is to carry out the DRM sedimentation in a weaker field, when the properties of any more strongly magnetized particles might be expected to show up. This was done with some representative samples using fields in the range 0·1–2·0 0e and the thermal demagnetization of the DRM carried out as before. No significant difference in the rate of decay of the DRM with temperature was observed whether the DRM was acquired in a 10 0e or a weaker field.

Another method of detecting different magnetic properties, in particular the presence of significant amounts of material with low or high blocking temperatures, is to examine the partial thermoremanence (PTRM) spectrum of the pigment and specularite. The intensity of PTRM acquired in any temperature interval can then be compared

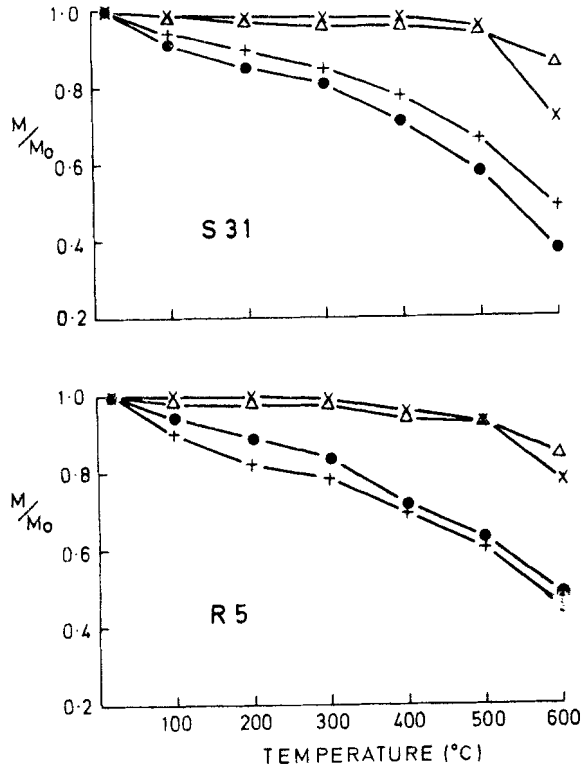

FIG. 3 Comparison of thermal demagnetization of DRM and PTRM spectrum of pigment and specularite fractions.
DRM: ×, specularite; ●, pigment. PTRM: △, specularite; +, pigment.

with the loss of intensity of DRM in the same interval during demagnetization; if the ratio of PTRM gained to DRM lost are similar in the same temperature interval, then there is evidence that the properties of the DRM are representative of the bulk of the particles in the two separates.

Samples of the pigment and specularite together with a test sample of the former were heated to successively higher temperatures in air (100°C intervals) and allowed to cool to room temperature in a field of 1 0e, the PTRM acquired in each interval being determined by subtraction. The IRM in 1000 0e of the test sample was measured after each heating to monitor any changes occurring, but no significant changes were observed. From the total TRM acquired by the samples the fractional loss during each temperature interval was determined and this compared with the fractional decay of DRM during thermal demagnetization. The results given by two samples are shown in Fig. 3. The two curves are very similar in each case, and it appears to be justified to assume that the NRM is carried by the specularite in these Group I samples, the pigment playing a negligible or only very minor role, since it does not appear to possess a high blocking temperature component.

The results shown in Figs 1 and 2 are from the Chugwater formation (R5, HC6, G3), Triassic, from the Western USA, Moenkopi formation (07), of the same age and from the same area, and the Supai formation (S31), Permian, and the Bonito Canyon quartzite (B17), Pre-Cambrian, both from Arizona. Other samples that fall into this group are from the Chinle formation (Triassic, Utah, USA) and from the Rio Chasquil area, Argentina (Triassic). Two samples of black haematite ore from Brazil also showed high stabilty of DRM.

Group II. This group consists of a variety of rock types and colours with widely

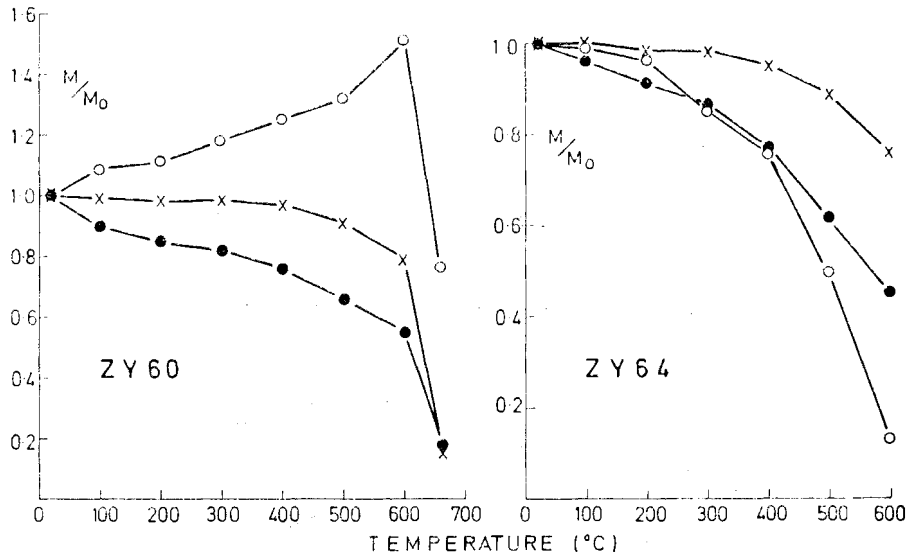

FIG. 4 Thermal demagnetization of NRM of the rock and DRM of pigment and specularite from samples of the Taiguati formation. Symbols as in Fig. 1

varying pigment/specularite ratios. There is sometimes not such a clear separation of pigment and specularite as in the Group I rocks. They are distinguished by the fact that the decay curve for the NRM of the rock is different from the DRM decay curves of both pigment and specularite. It appears possible to explain the results (Figs 4 and 5) on the basis that both forms of haematite contribute to the NRM of these rocks; in all the samples the specularite again possesses the hardest magnetization. The results

for samples ZY 60 and ZY 64 can be explained by the NRM of the pigment being reversed relative to the NRM of the specularite. In ZY 60 the NRM of the specularite is greater than that of the pigment and the more rapid decrease of the pigment NRM results in the NRM of the whole rock increasing until the temperature is near the Curie point. ZY 64 can be explained by assuming that the pigment NRM dominates in the rock and the more rapid decrease of the NRM of the rock at higher temperatures

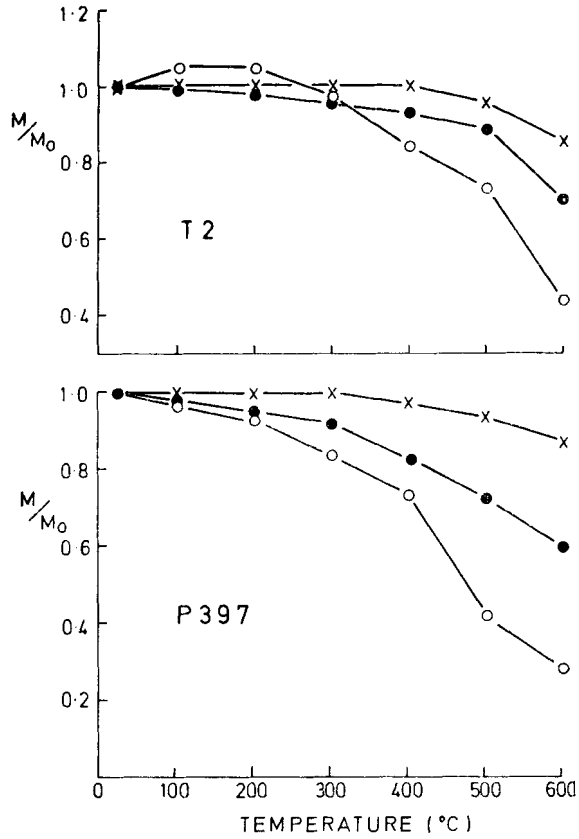

FIG. 5 Thermal demagnetization of NRM of the rock and DRM of pigment and specularite from two Group II samples.

is explained by the NRM of the rock being the difference between that of the pigment and specularite and therefore its proportional decrease can be greater than that of either of its components. This explanation can also be applied to P397, but T2 does not fit well into this category (see discussion). The rocks of this group are from the Brunswick beds (T2) of the Eastern USA, Triassic, the Taiguati formation, (ZY60, ZY64) Bolivia, Permo-Carboniferous, and the Picos beds (P397) Brazil, Devonian.

Group III. These rocks do not give clear-cut results. Like Group II, the separation of pigment and specularite is not well defined, and there is little difference between the DRM decay curves of the two haematite forms and the NRM of the rock. In the two examples shown, (Fig. 6) there is evidence of a small reversed component in GR13 but it is not possible to associate the NRM of the rock with either the pigment or specularite; in P403 there is some evidence that the NRM of the rock is associated with specularite, but this sample has a very large pigment content and only a small amount of specularite and it seems probable that the former must also contribute to the NRM.

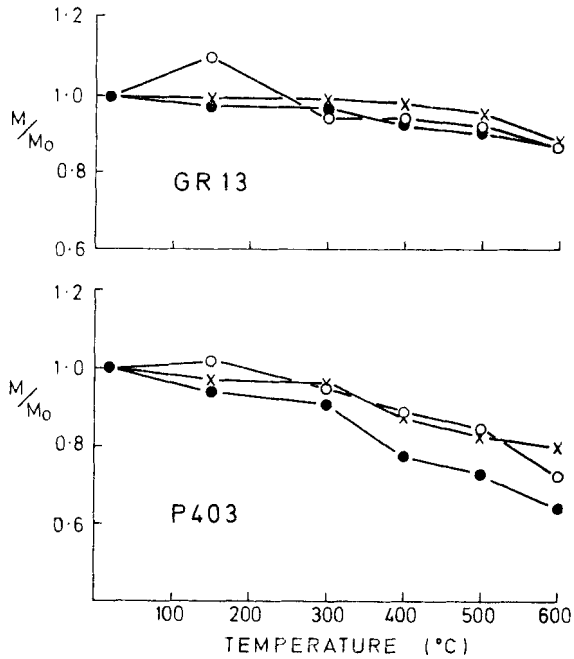

FIG. 6 Thermal demagnetization of NRM of the rock and DRM of pigment and specularite; Group III samples.

These rocks are from the Picos beds (P403) and the Grinnell formation (GR13), Montana, USA, Pre-Cambrian.

3.2 *The NRM of the specularite*

The DRM investigations afford a method of determining the average intensity of magnetization of the specularite particles. The first step is further investigation of the DRM intensity versus aligning field to examine whether complete alignment is achieved in a 10 0e field. If this is so, then analysis of the specularite fraction for haematite will enable its specific magnetization to be determined.

Small quantities (~30 mg) of the specularite fractions from rocks G1, G3, S31, ZY60 and ZY64 were deposited in fields of between 0·1 and 10 0e as previously described and the resulting DRM measured. Preliminary tests had shown that deposition

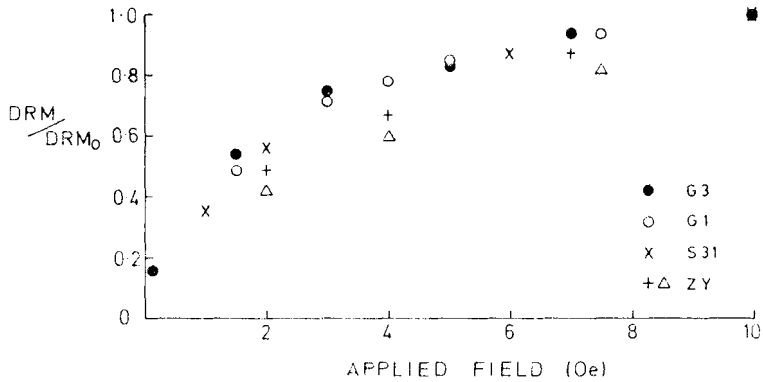

FIG. 7 Dependence of DRM intensity on applied field for specularite fractions.

in a particular field gave a DRM repeatable to about ± 10 per cent. The results are shown in Fig. 7, in which the intensity of DRM has been normalized to the value acquired in 10 0e. It is apparent that nearly complete alignment is achieved in 10 0e in G1, G3 and S31, and somewhat less in the ZY material. For comparison purposes the samples were also given a saturation IRM in a field of 10 k0e, and analysed for total iron as Fe_2O_3; four samples taken at random were also analysed for ferrous iron (as Fe0) which formed about 10 per cent of the total iron content; it probably occurs in ilmenite, which will also be extracted in the separation process, together with some clay mineral material which is also extracted. In Table 1 a nominal adjustment of this amount has been included in determining the specific NRM and IRM values. Since there may also be ferric iron present other than in haematite, the specific NRM and IRM values—quoted should be regarded as minimum ones.

Table 1.

DRM and saturated IRM intensities of specularite samples. Specific magnetizations are intensities per unit mass of specularite, based on ferric oxide content, as described in the text.

Rock	DRM (x 10^{-6}emu g^{-1})	IRM (x 10^{-3}emu g^{-1})	Specific magnetization (emu g^{-1}) DRM	IRM	IRM/NRM
G3	5000	43·0	0·020	0·17	8·6
G1	3800	33·4	0·017	0·15	8·8
G6	5200	56·5	0·018	0·19	10·9
HC2	4400	35·2	0·020	0·17	8·0
HC6	5000	28·2	0·029	0·17	5·6
R1	5500	32·0	0·031	0·18	5·8
R5	4100	20·0	0·023	0·11	4·9
R7	5500	39·0	0·025	0·18	7·1
O7	6500	38·7	0·036	0·22	6·0
NS1	—	—	0·048	0·17	3·6
NS2	—	—	0·041	0·17	4·1
ZY60	4600	37·5	0·030	0·24	8·2
ZY64	6300	33·3	0·033	0·18	5·3
S31	5300	54·0	0·020	0·20	10·1

A significant feature of these results is the low ratio of saturated IRM and NRM of the specularite in all cases, between about 4 and 10 in the rock-extracted material and even lower in the natural haematite ore (NS1 and NS2). Such a low ratio (~10) was also found by direct measurement of the NRM and saturated IRM of specularite grains extracted from samples of the Bonito Canyon quartzite (Collinson 1972).

4. Discussion

As a result of these investigations it is now possible to be more specific about the origin of the NRM of red sandstones. In all the samples examined it is the specularite which possesses the most stable magnetization; although the pigment always appears to be magnetized it is not aligned in some rocks and therefore does not contribute significantly to the NRM, but it is capable of imparting a DRM to a laboratory-deposited sediment. Only in the two Group III samples in the stability of the pigment DRM comparable with that of specularite. Because of the high coercivity of finely divided red ferric oxide it has hitherto often been considered that the observed high magnetic stability of many red beds is due to the red pigment content, but this now seems unlikely to be generally true.

The explanation proposed for the NRM of the Group I samples is as follows. The

remanence is a DRM carried by specularite particles, and comparison of the DRM acquired in a 10 0e field by the specularite with the NRM observed in the rocks shows that the particle magnetization directions are very scattered in the rocks. For instance, the NRM of sample G3 is $9{\cdot}0 \times 10^{-6}$ emu g^{-1}; the maximum DRM of the specularite from this sample, adjusted for its proportion in the rock (obtained from the weight of specularite extracted from a known amount of rock powder), is 108×10^{-6} emu g^{-1} and therefore the NRM is something less than 10 per cent of that obtaining if the particles were completely aligned. From the DRM versus applied field curve (Fig. 7) the NRM of this rock corresponds to an estimated applied field of about 0·1 0e which is at least in qualitative agreement with typical values of the Earth's field at present and in the past; however, it must be emphasized that applying the results of laboratory deposistion experiments to the original geological process is a difficult and uncertain procedure.

The pigment in the Group I rocks is magnetized but randomly directed in the rock and therefore does not contribute significantly to the NRM. It is tentatively concluded that the pigment was deposited with the rest of the sandstone material but was too weakly magnetized to align in the ambient field or was already attached to quartz grains; this latter feature would also inhibit the acquisition of a DRM through the magnetically 'dead' weight of the quartz grains. Considering sample G3 again, the intensity of DRM of the pigment is 16×10^{-6} emu g^{-1} in a 10 0e field and from the shape of the DRM intensity versus applied field curve the intensity in a 0·1 0e field is estimated to be about $0{\cdot}3 \times 10^{-6}$ emu g^{-1} ($\pm 30\%$) ,of the order of 3 per cent of the NRM of the rock; the uncertainty of extrapolating from laboratory to natural processes must be emphasized again, however.

Although the Group I formations are extensively described in the literature, there is little information available on the genesis of the pigment and specularite content. Baag & Helsley (1974) suggest, on the evidence of palaeomagnetic measurements on a long core of the Moenkopi formation, that the NRM was acquired simultaneously with deposition, consistent with a depositional origin of the magnetization.

In the Group II samples the pigment appears to be contributing to the NRM of the rocks and the suggested explanation of the results of thermal demagnetization (Figs 4 and 5) requires it to be reversely magnetized relative to the specularite; if this is so, then both forms of haematite cannot be contributing to the NRM through a contemporaneous DRM process. It is suggested that either the specularite first imparted a DRM and subsequently pigment was deposited from solution at a time when the field had reversed and it acquired a chemical remanence anti-parallel to the specularite component, or the pigment was deposited initially with the other rock material giving the sediment a DRM and then specularite was formed subsequently *in situ* and acquired a reversed field CRM. Of these alternatives the author favours the first. Neither the DRM intensity of the pigment or the absolute amount of pigment is large enough to suggest that a substantial DRM might be contributed to the rock by it, comparing the ZY samples with the pigment amounts and DRM intensity of the Group I samples. Also, the directions of the specularite particle magnetizations are clearly scattered again, based on the same evidence as for the Group I samples; this suggests a depositional rather than a chemical origin for the NRM due to the specularite.

While reversals are present in the Taiguati and Picos formations (Creer 1964), thus making the above interpretation feasible for these rocks, no reversals have been observed in the Brunswick beds (Collinson & Runcorn 1960; Opdyke 1961) although the initial increase of NRM during heating suggests the presence of a reversed component. These samples are a dark, purple-brown colour (near 5 RP 4/2 and 5 YR 4/1, Munsell colour chart) and contain much pigment and it seems probable that it is contributing much of the NRM in these rocks, and that reversed components are present which are not apparent in the palaeomagnetic directions.

It should be mentioned that a result such as that for P397 (and perhaps ZY 64)

could also be explained by incomplete separation of the specularite from the pigment, giving the latter an apparently greater stability, combined with a certain amount of experimental error in the shape of the demagnetization curves, However, this does not alter the important feature of the Group II samples, namely that pigment contributes an important part of their remanent magnetism.

Irving & Opdyke (1965), interpreting the palaeomagnetism and stability of the Bloomsburg red beds (Silurian), suggested that the observed NRM was carried by pigment and specularite, with the latter possessing the greater stability.

The results from the Group III rocks are difficult to interpret because of the similarity of the thermal decay curves of rock and separates. It seems likely that both pigment and specularite are contributing to the NRM of these samples.

5. Conclusions

The investigations reported in this paper provide evidence for a clearer picture of the origin of the remanent magnetism of sediments. Specularite, with a natural remanent intensity in the range 0·02 — 0·03 emu g^{-1} is the most important constituent in many formations, contributing a very stable magnetization of depositional origin. This component of NRM may or may not be associated with another component arising from chemical magnetization of pigment of post-depositional origin; there is evidence that detrital pigment deposited with the sediment does not contribute significantly to the NRM, although it is probably magnetized. The pigment almost always possesses a less stable magnetization than specularite, but sometimes exhibits comparable hardness. The wide variation in intensity of NRM in red sandstones is due to variation in the scatter of the magnetization axes of specularite particles within the rock, differences in specularite content and the presence or absence of a contribution from pigment, varying in intensity and possibly also in polarity. In an earlier paper on the NRM of red sediments the author (Collinson 1969) concluded that only a small proportion of the haematite present was giving the observed NRM; in the Group I sediments this appears to be true, but in other samples the low intensity of NRM (on which the conclusion was based) is apparently due to two anti-parallel magnetizations as well as directional scatter of particle magnetizations.

It is tentatively concluded from comparison of results from Group I and Group II and III rocks that if pigment is of post-depositional origin in a sediment it can acquire a chemical remanent magnetization of comparable intensity to the NRM arising from specularite. Deposition of detrital pigment results only in a very weak or negligible DRM which may not contribute significantly to the NRM of a rock in which specularite is present.

The technique described in this paper has not yet been systematically applied to red sandstones exhibiting unstable magnetization, it seems likely that the pigment carries the unstable component of NRM in these, and investigations on such rocks are being carried out. Although a reasonably representative group of rocks has been studied, the results are not necessarily true for all red sediments. Indirect evidence that pigment might be expected to be thermally harder than specularite has been given by Leng (1955) and Park (1970), although their conclusions were partly based on the properties of remanences other than NRM.

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