Geophys. J. Int. (2004) **159,** 909–916 doi: 10.1111/j.1365-246X.2004.02439.x

Laboratory chemical remanent magnetization in a natural claystone: a record of two magnetic polarities

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Accepted 2004 July 27. Received 2004 July 26; in original form 2003 May 7

SUMMARY

A record of two magnetic polarities during the acquisition by heating of a laboratory chemical remanent magnetization (CRM) in a natural pyrite-rich claystone was investigated. The samples were heated for 22 hr at 250 °C in a controlled magnetic field (1.0 mT) under an argon atmosphere. We interpret the origin of CRM as the surface oxidation of pyrite to magnetite, which in turn is progressively oxidized into haematite. We carried out experiments under a constant-polarity magnetic field and under two opposite polarities. The resulting CRM was measured after cooling in zero field: it is parallel to the applied field and has the direction of the last polarity. Thermal demagnetization under an argon atmosphere isolates an unexpected low-unblocking-temperature component ($T_{\mathrm{UB}} < 220$ °C). This component probably results from thermal alteration of magnetic carriers during subsequent thermal demagnetization. In the one-polarity experiments, thermal demagnetization of CRM above 220 °C isolates a well-defined component parallel to the imparted field direction. In the experiments with two magnetic polarities thermal demagnetization of CRM reveals two components of opposite polarity. The component which is parallel to the direction of the last applied field is well defined, while the other component, which has the polarity of the first applied field, is ill defined. Oppositely directed components are also detected by using alternating field demagnetization.

Key words: haematite, laboratory CRM, magnetite, pyrite-rich claystone, record of magnetic polarities.

INTRODUCTION

Remagnetization is a ubiquitous phenomenon in orogenic belts and their forelands (McCabe & Elmore 1989; Van der Voo 1993). The acquisition of a secondary chemical remanent magnetization (CRM) through the growth of new crystals of the magnetic carrier or the transformation of previous minerals is considered to be responsible for such remagnetization (Kligfield & Channell 1981; McCabe *et al.* 1989; Suk *et al.* 1990; Jackson *et al.* 1992; Banerjee *et al.* 1997; Enkin *et al.* 2000; Weil & Van der Voo 2002). However, the mechanisms for the remagnetization and their relationship to orogeny are not fully understood yet.

One controversial aspect of some orogenic remagnetizations is their single normal polarity (Hudson *et al.* 1989), especially for Tertiary remagnetization, because normal and reverse polarities of the Earth's magnetic field are to be equally expected (Morris & Robertson 1993; Villalain *et al.* 1994; Katz *et al.* 1998; Aubourg & Chabert-Pelline 1999; Katz *et al.* 2000; Henry *et al.* 2001; Cairanne *et al.* 2002; Kechra *et al.* 2003). The duration of the remagnetization process is still open to debate. Some authors propose a short time duration (less than 2 Myr) during a normal chron (Kechra *et al.* 2003). Conversely, Henry *et al.* (2001) propose a longer total duration for the acquisition of the magnetization. Actually, due to the chemical origin of the magnetization, the resulting CRM could be the sum of normal and reversed partial chemical magnetization components, as proposed by Merrill (1975), that would be carried by crystals with volumes that have exceeded the superparamagnetic single-domain threshold during normal or reverse magnetic polarities, respectively (the term 'partial' referring to a time interval in the course of the chemical process, *cf* O'Reilly 1984) Due to the probable overlap of their unblocking temperature spectra and of their unblocking field spectra, Henry *et al.* (2001) suggest that 'thermal and alternating field (AF) demagnetization methods may be inefficient at

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*Now at: CEREGE, Université Aix-Marseille III, Europôle Méditerranéen de l'Arbois, BP80, F-13545 Aix en Provence cedex 4, France. E-mail: cairanne@cerege.fr separating the normal and the reversed components', explaining why one may observe an apparent single component in a natural CRM, that is composed of partial CRMs of opposite polarities.

A recent experimental study of CRM carried by hydrothermally grown magnetite (Cairanne *et al.* 2003) demonstrates that laboratory grain-growth CRM can filter magnetic reversals, due to the overlapping unblocking field spectra of the partial CRM recorded by successive generations of magnetite. In this study, we propose an experimental test of the filtering of magnetic reversals in natural samples. We expect to produce a laboratory surface oxidation CRM by progressive replacement of previous minerals. With that aim we impart a magnetic field reversal during heating at a moderate temperature of natural sedimentary samples that have not been remagnetized. After cooling in zero magnetic field, we track two expected partial CRMs of opposite directions using thermal and AF demagnetization methods.

SAMPLES

We chose Upper Toarcian (Lower Jurassic) claystones from a borehole (Paris Basin, France) drilled by ANDRA (the French agency for nuclear waste). Those claystones constitute an unremagnetized equivalent of some remagnetized Jurassic rocks (Yang *et al.* 1996; Moreau & Ader 2000).

X-ray diffraction on powdered samples reveals the presence of clays (85 per cent), quartz (14 per cent) and pyrite (1 per cent) (Thiry & Schmitt 1992). All clay minerals (kaolinite, illite, chlorite, illite/smectite or chlorite/smectite) are of sedimentary origin or are of early diagenetic origin (Thiry & Schmitt 1992). Scanning electron microscope (SEM) study shows aggregates of framboidal pyrites of early origin, typically 5–20 μm in size, composed of individual crystals 0.5–1 μm in size.

EXPERIMENTAL PROCEDURE

Rock magnetism and microscopic observations

We carried out stepwise acquisition of isothermal remanent magnetization (IRM) and stepwise thermal demagnetization in air of a composite three-axis IRM (soft, 0.1 T; medium, 0.5 T; hard, 2.1 T) according to the method of Lowrie (1990). In order to detect subtle mineral alteration during heating, we used the method of Van Velzen & Zijderveld (1992). We monitored three pairs of samples to evidence different coercive fractions at 0.1, 0.5 and 2.1 T from 20 to 450 °C. One sample of each pair was subjected to IRM at 0.1, 0.5 and 2.1 T after each heating step, while the sister sample was only subjected to routine thermal demagnetization of IRM and served as a reference sample. We monitor both acquisition and demagnetization of the reinduced IRM. Samples were heated under an argon atmosphere. IRM was induced with a MMPM9 pulse magnetizer. Remanence measurements were performed with a DC SQUID magnetometer (2G-760R). SEM observations of rock powder were performed using a Hitachi S-2500, which is equipped with an energy dispersive spectroscopy (EDS) microanalysis device. The diameter of spatial resolution is less than 5 μm.

CRM experiments

To avoid the oxidized pyrite present at the surface of borehole cores, cylindrical samples of height 10 mm and diameter 10 mm were drilled just before each experiment. Heating experiments were conducted in a magnetically shielded non-inductive oven, under a controlled vertical magnetic field of 1.0 mT applied along the long axis of the core. An argon (99.997 per cent) atmosphere was maintained during the experiment. The temperature was stabilized for 15 min at 250 ± 1 °C in zero magnetic field. Then the vertical magnetic field was applied. When a magnetic reversal was imparted, the magnetic polarity was first downward and then upward. The sample was held at 250 °C for 22 hr, and then cooled in zero magnetic field in order to prevent partial thermoremanent magnetization between 250 °C and room temperature. Room temperature was reached after 1 hr. Circulation of argon was then stopped.

Components analysis

Samples were stepwise thermally demagnetized in an argon atmosphere in the same oven, or AF demagnetized up to 100 mT using a two-axis tumbler. We measured the remanent magnetization at room temperature using a SQUID magnetometer (CTF). Magnetic components were calculated by principal components analysis using Paleomac software (Cogné 2003).

RESULTS

Magnetic properties of the starting material

The starting material is characterized by a low natural remanent magnetization (NRM $< 3 \times 10^{-7}$ A m^{-2} kg^{-1}). Isothermal remanent magnetization (IRM) is saturated at 0.25 T (Fig. 1a). The stepwise thermal demagnetization in air of the three-axis IRM (Lowrie 1990) indicates a maximum unblocking temperature between 520 and 560 °C (Fig. 1b) for the soft component (0.1 T). The medium component (0.5 T) is small compared with soft component and the hard component (2.1 T) is negligible. Therefore magnetite constitutes the main original ferromagnetic carrier of the starting material.

Kinetics of the acquisition of laboratory remanent magnetization

One sample was heated to 250 °C in a downward 1.0 mT magnetic field in three successive runs: first for 6 hr, then for 16 and 38 additional hours. Remanent magnetization was monitored at room temperature after cooling in zero magnetic field. It always parallels the applied field, and its intensity increases logarithmically with cumulative time of heating (Fig. 2). After 22 hr of heating the intensity

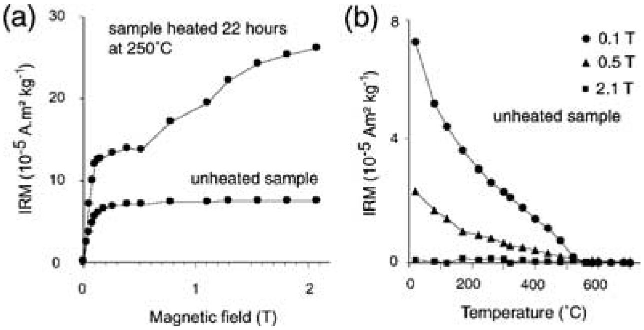


Figure 1. (a) Acquisition of IRM at room temperature for a representative unheated sample and for a representative heated sample. (b) Stepwise thermal demagnetization of a three-axis IRM imparted on a unheated sample, following the method of Lowrie (1990). Circles, triangles and squares correspond to soft (0.1 T), medium (0.5 T) and hard (2.1 T) magnetic components, respectively.

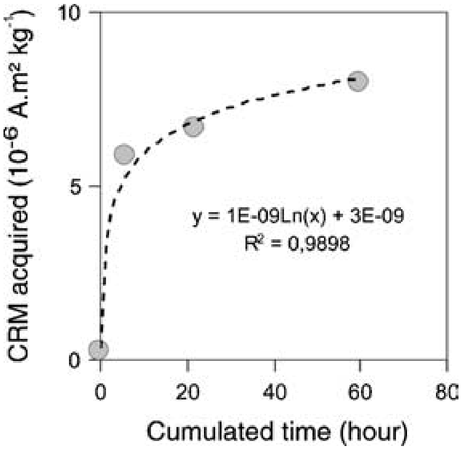


Figure 2. CRM acquired versus cumulative heating time at 250 °C for sample a1. The dashed line indicates the trend of change with time.

of the remanent magnetization is one order of magnitude higher than the NRM (Fig. 2).

Mineralogy and magnetic properties of the samples heated for 22 hr

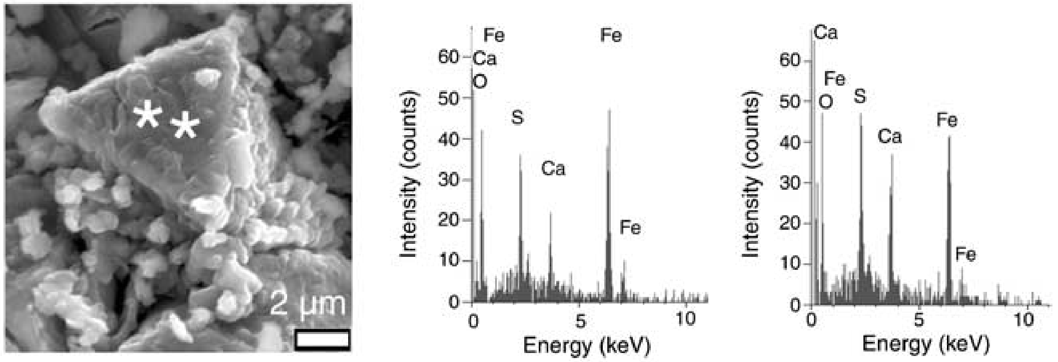
When heated for 22 hr at 250 °C and in an argon atmosphere, the colour of the samples changes from grey to light pink, and pyrite crystal aggregates of >1 mm in size are reddened. Moreover a sulphurous gas is released during the experiments. SEM observations of light pink rock powder (Fig. 3a) and of reddened pyrite aggregates (Fig. 3b) show in both cases crystals of 5–20 μm in size whose surface is composed of Fe, S, O and Ca.

The IRM acquisition curve of the samples heated for 22 hr indicates both soft- and hard-coercivity minerals: the soft one is saturated between 0.25 and 0.40 T, and the hard one is not yet saturated at 2.1 T (Fig. 1a). The method of Van Velzen & Zijderveld (1992) shows different behaviours between soft and medium components (Figs 4a and b) compared with the hard component (Fig. 4c). For all components the intensity of the IRMs reinduced at each step increases steadily from the first demagnetization step (80 °C) up to 360 °C. Above 360 °C, IRM intensities increase more rapidly. For soft and medium components (0.1 and 0.5 T), the IRM reinduced at each step is completely—or almost completely—demagnetized from 80 to 360 °C (Figs 4a and b). By contrast, the IRM reinduced at each step for the hard component (2.1 T) is not demagnetized and remains almost constant up to 360 °C (Fig. 4c). Above the 360 °C step, all IRMs reinduced at each step are not completely demagnetized and increase progressively. After demagnetization at 440 °C, 60 per cent of the high-coercivity component (2.1 T) remains, probably carried by haematite. The haematite is newly formed, as this mineral was not detected before heating. Rock magnetic measurements show therefore that haematite and magnetite are probably present when samples are heated for 22 hr at 250 °C under an argon atmosphere.

Magnetic components

We are aware that the magnetization acquired during these experiments is composite. It consists of a chemical magnetization produced by the growth of ferromagnetic grains through their

(a) matrix of the claystone



(b) reddened pyrite aggregate

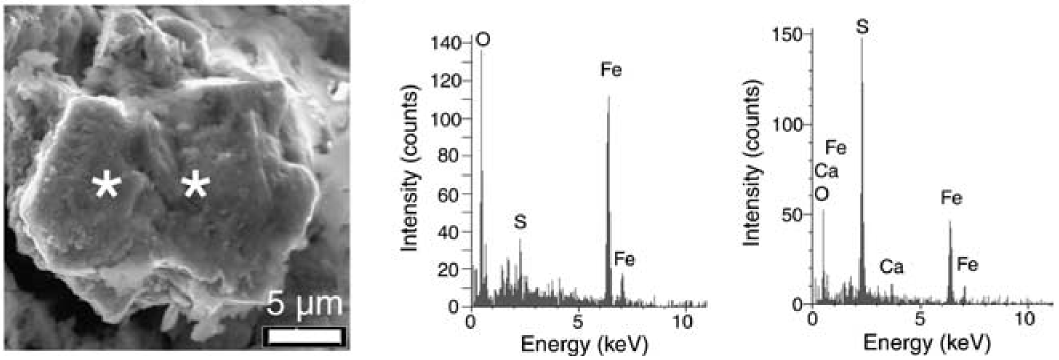


Figure 3. Microscopic observations on heated samples. (a) SEM image showing a pyrite crystal in the matrix of claystone; white stars locate the EDS analysis. (b) SEM image, showing a pyrite crystal in a reddened pyrite aggregate (>1 mm in size); white stars locate the EDS analysis.

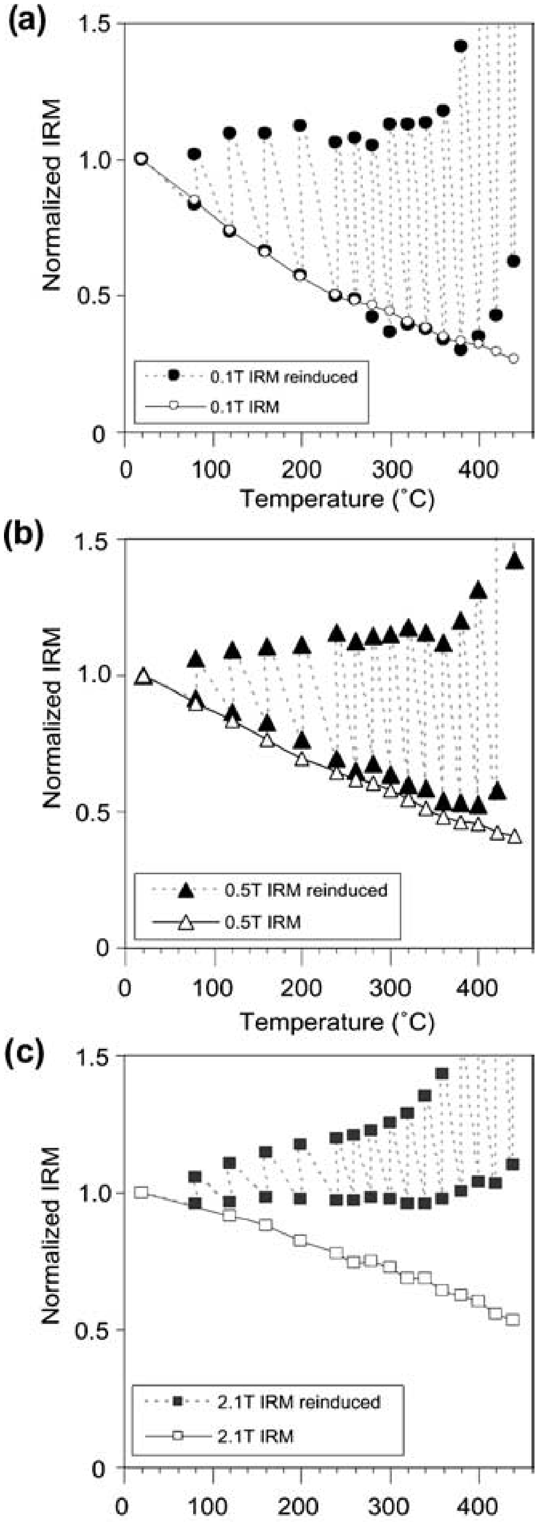


Figure 4. Results of thermal demagnetization under an argon atmosphere of paired samples after heating to 250 °C for 22 hr, using the method of Van Velzen & Zijderveld (1992) (see text for details). Circles, triangles and squares correspond respectively to 0.1 T IRM (a), 0.5 T IRM (b) and 2.1 T IRM (c). Open symbols correspond to IRM induced only once (reference sample) and full symbols correspond to IRM reinduced after each demagnetization step.

blocking volume and a thermoviscous magnetization produced by the applied magnetic field at a temperature of 250 °C for 22 hr maximum. Since the number and the volume of ferromagnetic grains are changing at each moment of the experiment, the chemical magnetization and the thermoviscous magnetization are not separable (Gie & Biquand 1988). The resulting magnetization is called chemical remanent magnetization or CRM.

Six experiments were performed: three with one polarity (upward or downward), and three with two polarities, first downward and then upward. The heating time was set at 22 hr for all experiments. We impart magnetic reversals either after 3.0 hr (∼14 per cent of the heating time, two experiments) or after 6.5 hr (∼30 per cent of the heating time, one experiment). According to the kinetics of the magnetic transformations (Fig. 2), this corresponds respectively to ∼50 per cent and ∼80 per cent of the CRM intensity acquired in one-polarity experiments. Assuming that the two expected partial CRMs add up, a null and a small downward resulting CRM are expected, respectively, for the two-polarity experiments. AF and thermal demagnetization results are shown in Figs 5 and 6 respectively. For clarity, declination is not represented in the Zijderveld diagrams in Fig. 5 and in Fig. 6 as inclination of CRM is very steep. Thermal demagnetization results are obscured by undesired mineralogical transformations above 45 °C, as suggested by the stepwise thermal demagnetization of IRM monitoring (Fig. 4).

Single-polarity experiments show a vertical CRM that parallels the applied field. AF demagnetization reveals one vertical component (Fig. 5a, Table 1). About 25 per cent of the CRM remains after the 100 mT demagnetization step. Thermal demagnetization shows an unexpected low-unblocking -temperature component (20–200 °C), called hereafter component A, that is directed oppositely to the applied field. It is followed above 200 °C by a vertical and well-defined component which is directed parallel to the applied field (Figs 6a and b, Table 1), called hereafter component B.

The two-polarity experiments show a vertical CRM that is directed parallel to the final applied field (Figs 6c and d). Given the kinetics of the magnetic transformation (Fig. 2), a magnetization parallel to the first applied field was expected in both cases (compare above).

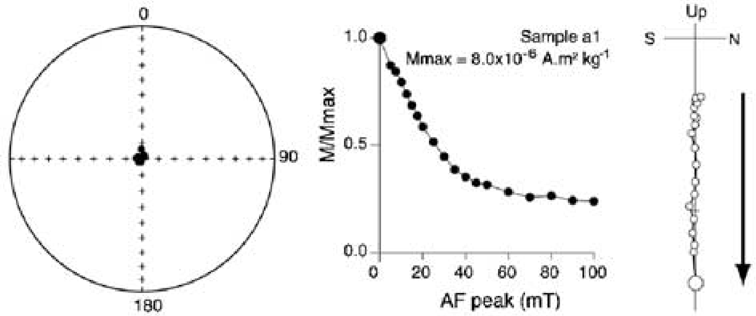
Reversal after 3.0 hr

AF demagnetization removes less than 50 per cent of the CRM. It reveals three nearly vertical components from 0–15, 15–40 and 50–70 mT (Fig. 5b, Table 1). The medium-unblocking-field component is directed parallel to the first applied field, while the low-unblocking-field component and the high-unblocking-field component are directed parallel to the final applied field (Table 1). Thermal demagnetization up to 450 °C removes ∼50 per cent of the CRM. Three components, A, B, C, are detected respectively between 20 and 120 °C, 120 and 300 °C and 300 and 450 °C (Fig. 6c, Table 1). These components are shown either by the negative slope of the variation of CRM intensity during thermal demagnetization or by the trend of inclination in the Zijderveld plot. Component B is vertical (see Table 1) and it is directed parallel to the final applied field. Although component A and component C are not vertical and poorly defined, their polarity corresponds to that of the first applied field. After 450 °C, a strong upward component remains. This component is called hereafter component D.

Reversal after 6.5 hr

Thermal demagnetization shows two components, B and C, above 200 °C (Fig. 6d). Note that component A is not observed here.

(a) no reversal applied, field down



(b) reversal applied after 3.0 hours

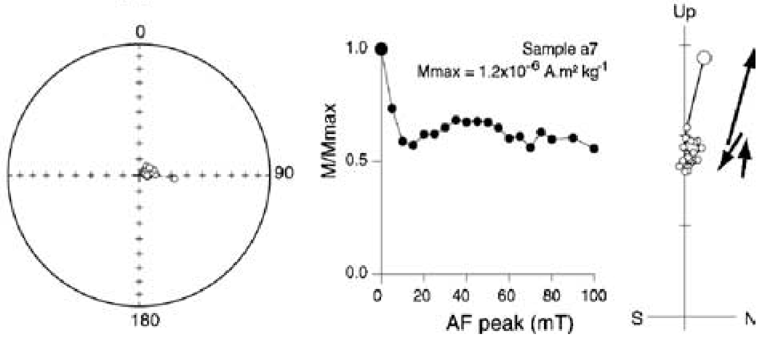


Figure 5. AF demagnetization results for experiments with one polarity (a) and two polarities (b): stereoplots, intensity plots, Zijderveld diagrams and their interpretation.

Component B (200 to 300 °C) is upward, vertical and well defined. Component C (300 to 440 °C) is not steep, downward and poorly defined (Table 1). Similarly to previous reversal experiments (reversal after 3 hr), component D remains above 440 °C.

In all cases, whenever the magnetic reversal is applied, component B is found to be nearly vertical and well defined ($\alpha_{95} \leq 8°$), while component A and component C are not vertical and are poorly defined (Table 1).

DISCUSSION

Alteration of the magnetic mineralogy during CRM acquisition

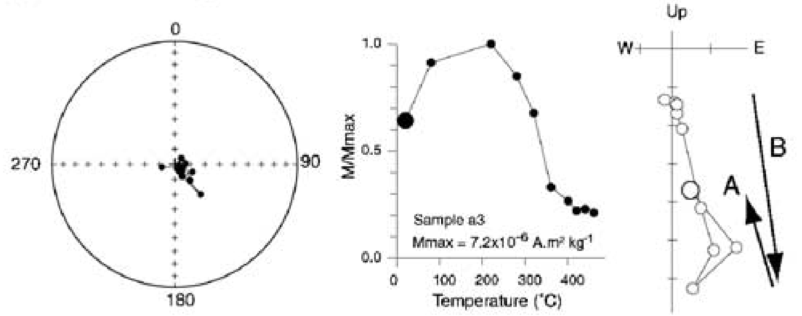
Magnetite is the only ferromagnetic mineral in the clay before heating. After heating, haematite and magnetite are present. We have no indication about the transformation of the initial magnetite during heating. Clay minerals (Hirt *et al.* 1993) and pyrite are two possible sources of iron. SEM data suggest that the pyrite has been oxidized in heated samples (Fig. 3) and that its surface is probably transformed into calcium sulphate and iron oxide/hydroxide/oxyhydroxide. Such secondary mineral phases are known to result from pyrite oxidation in mine tailings (Roussel *et al.* 1999) and in argilitic walls from tunnels (Charpentier *et al.* 2001). Moreover, oxidation rims of iron oxide/hydroxide/oxyhydroxide at the surface of the pyrite are frequently observed in sedimentary rocks affected by chemical remagnetization (Kligfield & Channell 1981; Suk *et al.* 1990; Banerjee *et al.* 1997), in weathered claystones (Crestin-Desjobert *et al.* 1988; Littke *et al.* 1991), in laboratory studies of pyrite oxidation (Nicholson *et al.* 1990; Nesbitt & Muir 1994; Descostes *et al.* 2002; Todd *et al.* 2003) and also in laboratory studies of CRM using pyrite as a starting material (Bina & Daly 1992).

We propose a tentative explanation for these results. Sulphides are present in noticeable proportion in our claystone (1 per cent of pyrite). On heating pyrite transforms into magnetite, by surface oxidation progressing from the pyrite surface toward its core, while magnetite is progressively oxidized into haematite. This hypothesis is supported by preliminary experiments performed on the same claystones heated at the same temperature range but in closed cells (Pozzi *et al.* 2003). In that case, only magnetite was produced. Thus haematite resulting from heating in an argon atmosphere is probably produced from the oxidation of magnetite. Therefore we interpret that the laboratory CRM is mainly acquired by surface oxidation of primary pyrite into rims of magnetite and haematite, leading to a CRM carried by a mixture of haematite and magnetite. Then the progressive increase of CRM would be the result of the cooperation of several physicochemical processes: an oxidation process that continuously alters the surface of pyrite into growing rims of magnetite and a gradual oxidation of magnetite into haematite. These reactions probably do not remain at a constant rate during heating but one can state that the final CRM is the sum of the magnetization of all the grains, magnetite and haematite, that grew beyond their blocking volume. The rate of production of haematite versus magnetite is not known. However, the fact that the latest polarity, which is also the longest polarity, is dominant in all reversal experiments supports a slow and continuous oxidation of magnetite into haematite.

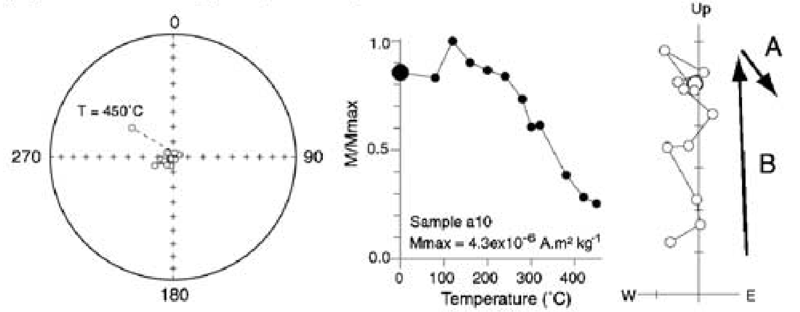
The origin of component A

Thermal demagnetization results display an unexpected feature below 120–220 °C. The first-cleaned magnetic component, component A, is directed nearly antiparallel to the CRM direction, even for experiments without magnetic reversal (Figs 6a and b). We did not expect a magnetic component with unblocking temperature lower

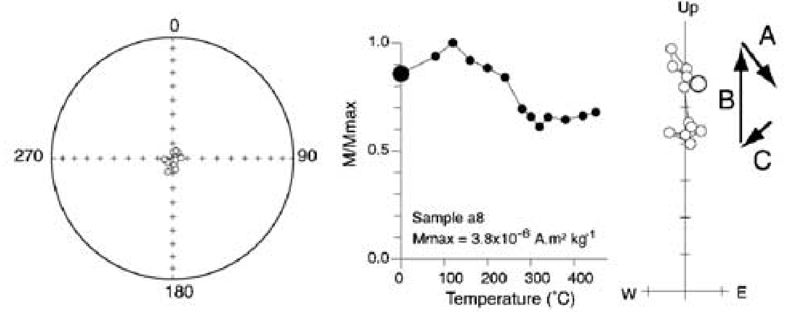
(a) no reversal applied, field down



(b) no reversal applied, field up



(c) reversal applied after 3.0 hours



(d) reversal applied after 6.5 hours

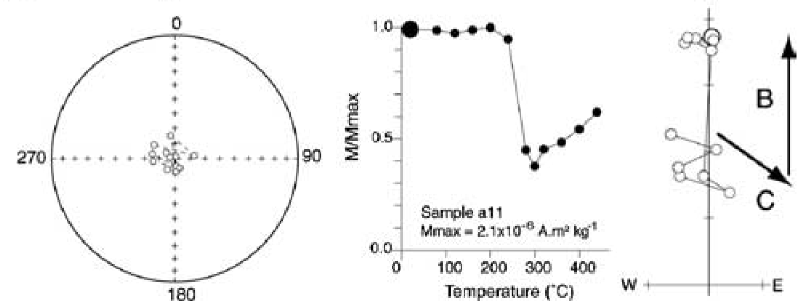


Figure 6. Results of thermal demagnetization under an argon atmosphere for experiments with one polarity (a and b) and experiments with two polarities (c and d). Same convention as for Fig. 5.

than 250 °C, because the sample is cooled to room temperature in a zero magnetic field. We checked that no CRM is carried by a sample heated for 22 hr at 250 °C in a zero magnetic field and then cooled to room temperature in a zero magnetic field. The increase in intensity of the CRM during thermal demagnetization between 20 and 220 °C could result either from the demagnetization of an antiparallel component, or from the creation of a parallel component. In the first hypothesis, component A could be interpreted as a self-reversed thermoremanent magnetization (TRM) acquired during cooling of heating experiments by fine crystals in the presence of the interaction field of the CRM-carrying grains. Self-reversed TRM acquired during cooling has been identified in several studies of laboratory CRM acquired by oxidation of ferromagnetic crystals (McClelland & Goss 1993; Bina & Daly 1994). In the second hypothesis, component A could be interpreted as the result of alteration of magnetic mineralogy during thermal demagnetization, leading to the creation of a magnetization that parallels the CRM direction. Results using the method of Van Velzen & Zijderveld (1992) show

Table 1. Table of magnetic components. Results are presented in the order of appearance in Figs 4 and 5. Components have been calculated using Paleomac software (Cogné 2003): *n*, number of data for calculation; $I°$, magnetic inclination in degrees; MAD, maximum angular deviation (Kirschvink 1980).

Sample	Demagnetization range	*n*	$I°$	MAD
a1	0–100 mT	18	+88	1
a7	0–15 mT	4	−77	10
a7	35–50 mT	4	+75	19
a7	50–70 mT	5	−84	2
a3	20–220 °C	4	−29	23
a3	220–440 °C	7	+80	5
a10	20–120 °C	3	+50	12
a10	120–440 °C	9	−86	5
a8	20–120 °C	3	+26	25
a8	120–320 °C	7	−87	3
a8	320–450 °C	5	+23	40
a11	200–300 °C	4	−85	8
a11	300–440 °C	5	+37	35

that magnetic carriers are created as early as the first step of demagnetization of the heated samples (Fig. 4). The hard fraction of these new magnetic carriers is created below their unblocking temperature, thus contributing to remanence. These results are in favour of the second hypothesis. We suggest that component A results from the production of new magnetic carriers during thermal demagnetization, probably by crystallization of poorly crystallized material at the surface of CRM-carrying grains.

Chemical record of magnetic polarities

Besides this low-temperature component, the prominent result reported here is that at least two components are found for the two-polarity experiments, using AF or thermal demagnetization. Thermal demagnetization shows upward components (B and D) and a downward component (C) that could correspond respectively to the second applied polarity and to the first applied polarity. We interpret component B and component D as the record of the second applied field. We interpret component C as the record of the first applied field because component C is not isolated for the single-polarity experiments (Figs 6a and b).

We propose the following scenario. Before the reversal, a downward remanent magnetization is acquired by newly formed magnetic carriers as they grow through their blocking volume, and by previously existing carriers (detrital magnetite). After the reversal, upward remanent magnetizations (component B and D) result from reorientation of some fraction of the downward remanent magnetization, that may be stabilized by the growth of the carriers, and growth of newly formed magnetic carriers as they pass their blocking volume in the upward field. The remaining fraction of the downward remanence corresponds to component C.

Implication for natural CRM

The components of natural chemical remagnetization are often carried by magnetite in carbonates or by haematite in sandstones (McCabe & Elmore 1989). However, several studies describe remagnetization of carbonates carried by haematite (McCabe & Elmore 1989), or carried by both haematite and magnetite (Henry *et al.* 2001). The remagnetization in sediments has been ascribed to a CRM carried by secondary ferromagnetic minerals that replace pre-existing pyrite in several studies (Kligfield & Channell 1981; Johnson *et al.* 1984; Suk *et al.* 1990; Banerjee *et al.* 1997). From that point of view, we believe that our laboratory experiments simulate such a mechanism of remagnetization.

The experimental chemical remagnetizations reported in this study are probably the result of several physicochemical processes where magnetite partly replaces pyrite and is then oxidized into haematite. In the case of natural surface oxidation CRM during two magnetic polarities, leading to a mixture of magnetite and haematite, our results suggest that two partial CRMs of opposite polarities may be separated due to the juxtaposition of their unblocking fields and temperature spectra.

Our results contrast with the results obtained with CRM resulting from nucleation and growth of hydrothermal magnetite (Cairanne *et al.* 2003). We know that in that case only magnetite is formed and that partial CRMs of opposite polarities are not separable, due to the perfect superposition of their unblocking field spectra. We propose that the separation of the partial CRMs in the present study is possible because the chemical process is the surface oxidation of pyrite. Further study is needed to quantify the influence of the subsequent alteration of magnetite into haematite. Our results, together with those of Cairanne *et al.* (2003), bring new elements to the understanding of pervasive remagnetization with a unique polarity.

CONCLUSION

We produced laboratory CRM while heating pyrite-rich natural claystones at 250 °C, under an argon atmosphere, and in a controlled 1.0 mT magnetic field. The main results are as follows:

(1) CRM results mainly from the alteration of pyrite into a mixture of magnetite and haematite. The progressive increase of CRM is probably a result of a combination of an oxidation process that continuously alters the surface of pyrite into growing rims of magnetite and a gradual oxidation of magnetite into haematite.

(2) For two-polarity experiments, CRM parallels the last applied field, and it is possible to separate the components of opposite polarities.

(3) Thermal demagnetization under an argon atmosphere isolates four magnetic components. Component A, removed below 220 °C, is unexpected and probably results from alteration of magnetic carriers during thermal demagnetization. We interpret the other three components as a complex record of the two polarities. The well-defined component B is parallel to the second applied field. We interpret it together with component D as a good-quality record of the second applied field during laboratory CRM acquisition. The poorly defined component C has the polarity of the first applied field.

ACKNOWLEDGMENTS

This work was funded by CRNS. ANDRA (the French agency for nuclear waste) provided samples and an unpublished mineralogical study. The authors wish to thank F. Brunet and P. Rochette for fruitful discussions and M. Jackson and an anonymous reviewer for their careful and constructive reviews. H. Soffel is gratefully acknowledged for his handling of the manuscript.

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