Geophys. J. R. astr. Soc. (1983) 74, 667-687

Viscous magnetization of 0.04–100 μm magnetites

David J. Dunlop[*] *Université de Paris VI et CNRS, Laboratoire de Géomagnetisme, 94100 St-Maur-des-Fossés, France*

Received 1982 December 12; in original form 1982 September 23

Summary. Acquisition and decay of small-field viscous magnetization have been measured as a function of temperature (9–500°C) for four synthetic magnetite dispersions ranging in mean particle size from 0.04 to 0.22 mkm and at room temperature only for an additional six samples (2–100 mkm). Room-temperature viscous magnetization is pronounced in the single-domain range, slight between 0.1 and 5 μm, and again substantial in the intermediate multi-domain range (10–15 mkm). In all experiments, the acquisition rate exceeded the decay rate, resulting in residual undecayed viscous remanent magnetization (VRM) after a zero-field decay time equal to the time of exposure to a field. Viscous magnetization is enhanced at elevated temperatures but not in direct proportion to absolute temperature. Instead, there appears to be a low-temperature (< 20°C) threshold below which viscous effects are negligible and an approach to saturation at high temperature. VRM produced isothermally at a given temperature is significantly more difficult to erase by heating to a higher temperature than most thermal activation theories predict. Furthermore, even in quite brief experiments, the viscosity coefficient $S = J/\partial \log t$ increased with time, the increase being more pronounced at high temperature. The implications of these observations are that viscous overprinting of natural remanent magnetization in magnetite-bearing rocks, particularly during burial or intrusive reheating, may be more extensive and more difficult to erase than previously believed.

Introduction

Following any change in applied magnetic field, the magnetization of a ferrimagnetic substance relaxes towards a new thermal equilibrium state. Stable remanent magnetization is a consequence of the extremely long relaxation times of relatively large single-domain (SD) grains or of strongly pinned domain walls in multidomain grains. However, there will always be a fraction of either SD moments or pinned domain walls that relax during the time span of an experiment.

[*] Permanent address: Geophysics Laboratory, Department of Physics and Erindale College, University of Toronto, Toronto M5S 1A7, Canada.

The time-scale of interest may range from the few seconds or minutes required to measure the natural remanent magnetization or NRM of a sample (e.g. Urrutia-Fucugauchi 1981) to the length of the Brunhes epoch, during which the viscous remanence or VRM component of NRM has been acquired (e.g. Dunlop & Hale 1977). Even longer time-scales are possible if a rock suffered long exposure to a field of one polarity during burial or intrusive heating and the resulting viscous partial thermoremanence or VPTRM was then stabilized by cooling (e.g. Briden 1965). VRM of recent origin constitutes noise, since it records only the present field and tends to obscure primary NRM of more ancient origin. VPTRM, on the other hand, is a source of valuable palaeomagnetic and tectonic information if the time and temperature of heating can be determined.

Viscous magnetization (also called magnetic viscosity or magnetic after-effect) has been the subject of a great many studies, the most notable being the theories of Preisach (1935), Richter (1937), and Néel (1949, 1950) and the experimental work of Street & Woolley (1949), Barbier (1953), Creer (1957), Le Borgne (1960) and Shimizu (1960). (For a review of the relevant literature, see Dunlop 1973a). Despite the wealth of information available, many aspects of viscous magnetization remain in doubt or even in dispute. For example, the idea that VRM is acquired or decays in proportion to the logarithm of elapsed time, which is the justification for defining a viscosity coefficient $S = dJ_{\mathrm{vrm}}/d\log t$, is firmly rooted in the literature and dates back at least to the work of Thellier (1938). However, although $\log t$ viscous changes are frequently observed, non-logarithmic changes are also common (e.g. Lowrie & Kent 1978), particularly with materials that are strongly viscous. It is not clear whether two distinct classes of materials are involved or the same phenomenon viewed at different magnifications, as it were.

Viscous effects are inevitably difficult to categorize in any systematic fashion because they are a manifestation of the properties of a narrow range of SD particle sizes or domain wall pinning characteristics. In most experimental samples, only a small fraction of the magnetic domains are viscous. Even in carefully sized samples it is difficult to know — as many authors have noted — whether the effects observed are inherent or due to ‘contamination’ by a minute fraction of ultrafine particles.

Among previous studies, only Shimizu (1960), Zhilaeva & Minibaev (1965) and Tivey & Johnson (1981, 1982) have attempted to study the intrinsic grain-size dependence of magnetic viscosity in the palaeomagnetically important mineral, magnetite. Shimizu reported little difference in viscosity coefficent 5 between 2 and 100 mkm grains, Tivey & Johnson (1982) found that > 45 mkm grains had S almost three times as large as 10 mkm $< d <$ 45 mkm grains, while Zhilaeva & Minibaev reported that S was weakly size-dependent between 20 and 100 mkm but increased four-fold with *decreasing* particle size between 2 and 20 mkm.

The present study attempts to clarify this contradictory experimental evidence by using magnetites of a common provenance that are narrowly sized in individual samples but collectively span the broadest size range possible. Four samples contained single-crystal cubes precipitated in aqueous solution with mean sizes in the range 0.04–0.22 mkm and size dispersions of 20–40 per cent about the mean. Six samples, kindly supplied by Dr M. E. Bailey, University of Toronto, contained irregular particles crushed from large (> 150 mkm) single crystals and separated into size fractions using a Bahco dust analyser. Mean sizes ranged from 2 to 100 mkm with much greater size dispersion than in the submicroscopic samples and the very real possibility of adhering fine particles (*cf.* Parry 1965). These large samples are nevertheless probably superior to those of previous workers, who used fairly coarse mesh separations.

The results of this study are complementary to those of Tivey & Johnson (1981, 1982), who examined prinicipally the effects of grain size, concentration, initial state and storage time on room-temperature viscosity. The present results generally confirm Tivey & Johnson's data for the size dependence of viscosity in submicron grains, considerably augment their data base in the > 1 μm range, and focus attention on the thermal and isothermal behaviour of viscous magnetization at high temperatures. This latter subject is vital to understanding and calibrating thermoviscous remagnetization (i.e. acquisition of VPTRM) in nature.

Theoretical background

Viscous magnetization (VM) is the time-dependent change, in magnitude and direction, of the prior magnetization **J** of a sample as a result of a change in applied magnetic field $\mathbf{H}_0$ occurring at time $t = 0$. If $\mathbf{H}_0$ is suppressed at time t, the remanence will consist of: (1) any surviving prior remanence; (2) isothermal remanence or IRM acquired quasi-instantaneously when H_0 was changed; and (3) VRM, $\mathbf{J}_{\mathrm{vrm}}(\mathbf{H}_0, t)$. Note that VM is measured with the field on, while VRM is measured with the field off. For weak fields ($H_0 < 10$ Oe), it is generally observed that IRM $\propto H_0^2$ whereas VM and VRM $\propto H_0$. Thus the importance of VM and VRM relative to IRM increases with decreasing field strength.

An ensemble of SD particles, each with volume V and anisotropy constant (shape or crystalline) K, undergoes magnetization relaxation described by

$$\mathbf{J}(t) - \mathbf{J}_{\mathrm{eq}} = [\mathbf{J}(0) - \mathbf{J}_{\mathrm{eq}}] \exp(-t/\tau) \tag{1}$$

where $\mathbf{J}_{\mathrm{eq}} = \mathbf{J}(\infty) \parallel \mathbf{H}_0$ is the thermal equilibrium magnetization for field $\mathbf{H}_0$ and temperature T. The relaxation time τ, for small $\mathbf{H}_0$, is given by

$$\tau^{-1} = f_0 \exp[-VK(T)/kT],$$

or (2)

$$\log\tau = -\log f_0 + VK(T)/kT$$

where k is Boltzmann's constant and f_0 is a frequency $\approx 10^{10}\,\mathrm{s}^{-1}$.

Using a magnetization blocking approximation ($\mathbf{J} - \mathbf{J}_{\mathrm{eq}} = \mathbf{J}_0 - \mathbf{J}_{\mathrm{eq}}$ if $t \lesssim \tau$, $\mathbf{J} - \mathbf{J}_{\mathrm{eq}} = 0$ if $t \gtrsim \tau$) instead of (1) and assuming a uniform distribution $f(\log\tau)$ of SD relaxation times, Néel (1949) showed that

$$|\mathbf{J}(t) - \mathbf{J}(0)| = J_{\mathrm{vm}} = J_{\mathrm{vrm}} = S_{\mathrm{a}} \log t \tag{3}$$

or that if $H_0 = 0$,

$$|J_{\mathrm{vrm}}(t) - J_{\mathrm{vrm}}(0)| = S_{\mathrm{d}} \log t. \tag{4}$$

S_{a} and S_{d}, the coefficients of viscous acquisition and decay of magnetization, should be equal and S_{a} or S_{d} are predicted to vary with temperature as $T/K(T)$. The results (3) and (4) are a direct reflection of the assumed uniform distribution $f(\log\tau)$ and are largely independent of the decay law for individual ensembles (see Dunlop 1973a, p. 863).

Thermal activation of pinned domain walls follows a law analogous to (2) (Gaunt 1976; Gaunt & Mylvaganam 1979), with V now referring to the volume involved in a single Barkhausen jump and K being a measure of pinning energy. The frequency constant f_0 is in the range $3 \times 10^9\,\mathrm{s}^{-1} \lesssim f_0 \lesssim 10^{12}\,\mathrm{s}^{-1}$ (Gaunt 1977) so that $\log f_0$ is not substantially different from its SD value. We may anticipate, therefore, that the laws of acquisition and decay of viscous magnetization will be mathematically similar in SD and MD grains, although the viscosity coefficients S_{a} and S_{d} may be of quite different magnitudes.

In fact, Street & Woolley (1949) treat thermal fluctuations in the heights of energy barriers opposing wall motion (the pinning energy) as analogous to fluctuations in barriers to rotation of SD moments and derive results equivalent to (3) and (4) for MD grains. In their theory, S_a and $S_d \propto T/J_s(T)$ where $J_s(T)$ is spontaneous magnetization.

On the other hand, Néel (1950) takes the point of view of a fluctuating internal viscosity field that imparts energy to domain walls. He finds, for weak fields,

$$J_{\mathrm{vrm}}(t) + J_{\mathrm{irm}} = \tfrac{1}{2}p\,(H_0 + S_v \log t)^2 \tag{5}$$

in which J_{irm} is IRM, p is a constant related to the second Rayleigh coefficient and S_v is the viscosity field coefficient (*cf.* Dunlop 1973a, p. 872). Depending on the relative magnitudes of H_0 and $S_v \log t$, the VRM may go as $\log t$, as $(\log t)^2$ or as a quadratic function, $a \log t + b (\log t)^2$. $S_v \propto T^{1/2}$ is predicted, for T far from the Curie temperature. Furthermore, S_a for VM acquisition should be twice S_a or S_d for VRM acquisition or decay.

A theory recently proposed by Walton (1980) for SD grains, and perhaps adaptable to MD wall displacements as well, predicts

$$J_{\mathrm{vrm}}(t) = CH_0\,[T/K(T)]^{2+r}\,(\log f_0 t)^{3+r} \tag{6}$$

where C is a constant and r is a power-law index in the distribution function $g(V) \propto V^r$ [or the equivalent distribution $f(\log \tau) \propto (\log \tau)^r$]. Stephenson (1971, equation 9) has derived a similar, slightly more complex expression. In these theories, the time and temperature dependences of magnetic viscosity are correlated and both depend on the distribution $f(\log \tau)$. For a uniform distribution, as assumed by Néel (1949) and Richter (1937), $r = 0$ and $J_{\mathrm{vrm}} \propto [T/K(T)]^2 (\log f_0 t)^3$.

Experimental procedure

The instrument used in VM and VRM experiments was an astatic magnetometer system with spot-follower amplifier and continuous strip-chart recording (Plessard 1971). Transients died away and attenuation and DC offset adjustments (the latter to compensate induced magnetization IM or IRM) could be completed within ~ 4 s of changing the applied field. The first data point was taken from the recording at 5 s, the last at 20 min, and 12 intermediate values at approximately equal increments of log *t*. Although the record is short, it nevertheless covers 2½ decades of *t* ($0.7 < \log_{10} t < 3.1$). Little would have been gained by extending the record to 30 or 40 min, especially as drift due to temperature changes in the uninsulated experimental hut became significant over longer periods.

Samples (1.6 cm diameter x 5 cm long cylinders) were mounted in a non-inductive vacuum furnace adjacent to the magnetometer. A coaxial solenoid applied an axial field to the sample, and changes in the axial magnetization were detected by the magnetometer. A standard field of 10 Oe was used in most runs. Helmholtz coils nulled the ambient Earth's field during decay experiments and between runs at different temperatures.

Samples were AF demagnetized before initial and final room-temperature runs but only rarely between high-temperature runs. Instead thermal demagnetization was used. Following a 20 min VM run and a 20 min VRM decay run at a given temperature, the sample was heated in zero field to the next highest temperature and left for a wait time of about 30 min. Above 300°C, the temperature steps were only 50°C and wait times $\gtrsim$ 1 hr were sometimes required, particularly at 350 and 400°C. Experience showed that rather different viscosity coefficients resulted if the remanence from the preceding run was not fully erased. If for any reason (noise, drift, unsuitable attenuation or offset), a run had to be abandoned or the

Preparation and p

results rejected, the run could not be repeated immediately but only after cooling and repeating the initial sequence of AF demagnetization, pumping down and heating.

The experiments differ in two ways from many previous studies. First, viscous acquisition was monitored continuously in the presence of the applied field, so that induced magnetization (VM) rather than remanence (VRM) was measured. Tivey & Johnson (1981, 1982), who used the same procedure, point out its advantages in saving of time and continuity of record. Secondly, room-temperature experiments used an AF demagnetized initial state, whereas all other runs started from a partially thermally demagnetized condition.

Preparation and properties of samples

The ten experimental samples were dilute dispersions of magnetite powders of various controlled sizes, high purity kaolin forming the matrix and cement. Experience has shown kaolin to be perfectly inert and non-reactive with magnetite at all temperatures, provided the sample is slowly heated in vacuum to ≈ 350°C to completely dehydrate the kaolin. Five anneals for a total time of 16½ hr were used. No further thermal stabilization was carried out because at temperatures ≳ 600°C the finer magnetite grains, which are inevitably clumped to some degree, tend to sinter. One is then measuring the properties of material with a different effective grain size and probably a more complicated domain structure. Of course, there is a danger that such changes may begin to occur even at 500°C, the highest temperature used in VM runs. Room-temperature hysteresis properties, initial susceptibility and viscosity coefficients measured before and after the high-temperature runs indicated no systematic physical or microstructural changes, however.

Four samples used a series of submicron magnetites that have been used in a number of previous studies. The properties of these powders have been described in detail elsewhere (Dunlop 1973b). The particles are single-crystal cubes precipitated from $FeSO_4$ and, like ultrafine natural magnetites, slightly surface oxidized. Dr P. R. Buseck of Arizona State University, who has examined the powders under the transmission electron microscope, considers them to be 'remarkable in their degree of crystallinity and uniformity of size',

Table 1. Grain size of the magnetite powders and hysteresis properties of experimental samples made from them.

Grain size (μm)						
Mean	Sd or range	$(\chi_0)_i^*$ (emu cm^{-3} Oe^{-1})	$(\chi_0)_f^*$	$H_c^\dagger$ (Oe)	$H_R/H_c^\dagger$	$J_r/J_s^\dagger$
0.037	±0.015	0.150	0.153	175	1.94	0.254
0.076	0.025	0.154	0.152	180	1.75	0.270
0.100	0.029	0.157	0.156	165	1.82	0.262
0.221	0.042	0.184	0.085	135	2.15	0.174
2.1	1.2-3.2	(0.153)		145	1.58	0.26
3.8	2.2-5.4	(0.203)		95	1.74	0.22
6.2	3.7-7.0	(0.261)		70	1.93	0.16
9.8	6.3-13.3	(0.207)		47	2.45	0.09
14.3	7.4-15.5	(0.279)		45	2.56	0.08
	100-125	(0.242)		20	8.6	0.015

*Values of RT initial susceptibility χ_0 are per cm^3 of magnetite. Values for the last six samples are less certain because the magnetite content is not known precisely. Subscripts i, f mean before and after all VM/VRM runs respectively.

†H_c, coercive force; H_R, remanent coercive force; J_r, J_s are near-saturation IRM and IM. All determinations were made using a maximum field of 1.5 kOe.

(1980, private communication). The four powders are similar in all respects except mean grain size, which ranges from 0.04 to 0.22mkm(Table 1). Size dispersion is sufficiently narrow that there is minimal overlap between size ranges of different powders.

The 6mkmsize and larger powders were prepared by ball-milling large single crystals, followed by dust separation. Their properties are described fully by Bailey & Dunlop (1983). These magnetites are more irregular in form and have more mutual size overlap than the submicron series. The size spreads quoted in Table 1 are 90 per cent cumulative volume limits.

The volume concentration of the submicron magnetites was 1 per cent. The micron-size magnetites had previously been dispersed in powdered silica. After mixing with kaolin in 1:3 proportions, the magnetite concentration was about 0.25 per cent by volume. Precise VM measurements were correspondingly more difficult with this series.

Initial susceptibility χ_0 (the only bulk property that could be monitored at each heating step) showed that the 0.22 mkm sample underwent oxidation at 455 and 500°C for reasons that are unclear (a vacuum leak?). After cooling, χ_0 was less than half its pre-heating value (Table 1). The other submicron samples did not change chemically. However, unexpectedly, the micron-size series all suffered significant oxidation. Only room-temperature VM results are reported for this series.

Room-temperature hysteresis was measured with a 1. kOe solenoid and multi-stroke ballistic magnetometer designed by A. Lecaille. The parameters listed in Table 1 were measured after 350°C annealing but before VM/VRM runs. Values measured after the 500°C run were generally similar.

Hysteresis parameters of the submicron series are those expected for small MD grains approaching SD size: $_c \gtrsim 100$ Oe; H_R/H_c 2; $_r/J_s > 0.15$. In the > 1mkmseries, x_0 increases in a general way with increasing grain size, but H_R/H_c and J_r/J_s for the 2.1, 3.8 and 6.2mkmsamples are similar to submicron values. Adhering submicron particles could hardly bias bulk hysteresis to this extent (although they could considerably bias viscous behaviour). Internal strain and irregular shape likely account for the hard hysteresis properties, these being the powders that were ball-milled the longest. The 9.8, 14.3 and ≈ 100mkmsamples have hysteresis parameters compatible with Parry's (1965) measurements.

Experimental results

GRAIN SIZE DEPENDENCE OF VM AND VRM

Measured room-temperature values of S_a and S_d are given in Fig. 1. Since VM and VRM changed in a non-logarithmic fashion with time in these experiments, the values shown are averages $S_a = \Delta J/\Delta \log t$, $S_d = |\Delta J_r/\Delta \log t|$ over a 20 *min* experiment, not instantaneous values $\partial J/\partial \log t$, $|\partial J_r/\partial \log t|$. VM and VRM were reduced to values appropriate to 100 per cent magnetite and $H_0 = 1$ Oe. To make the latter reduction, it was necessary to assume J, $J_r \propto H_0$. This assumed field dependence was approximately verified for one submicron and one > 1 μm sample at room temperature. It was not checked in a systematic way for other samples or at higher temperatures.

The S_a data are generally consistent with values from the literature (points S2, S3, S4 from Shimizu 1960; Z, LR, CC, MAG 5 from Tivey & Johnson 1981, 1983). A possible exception is S_a for the ~ 100 mkm sample, which is 1.5–2 times less than previous values for similar crushed material. Gaps in the previous data set, from 0.02–0.1 mkm and 2–100 μm, are partially bridged by the new results.

A sharp decrease in S_a between small, nearly superparamagnetic SD grains (0.02–0.04 mkm) and grains near or just above critical SD size (0.05–0.2 mkm) is confirmed. Such a decrease is

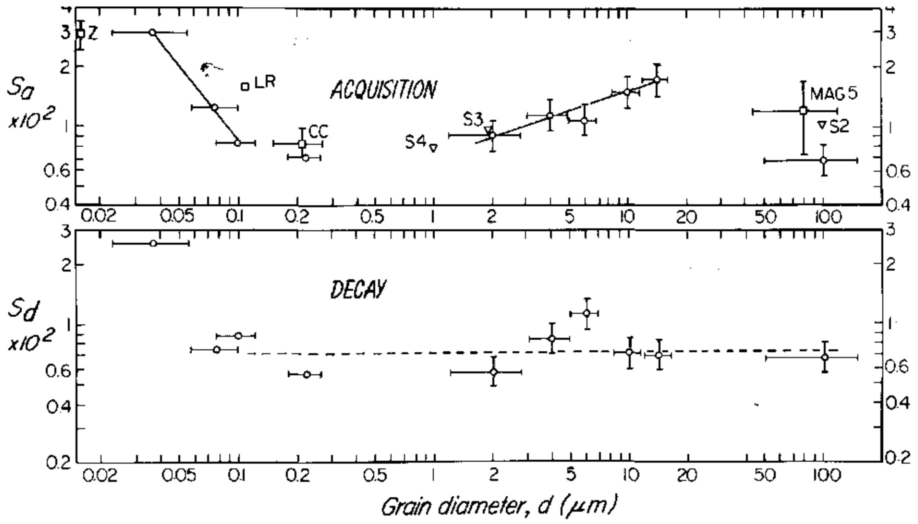

Figure 1. Average room-temperature viscosity coefficients S_a (VM acquisition) and S_d (VRM decay) in units of emu cm^{-3} for grains of magnetite as a function of grain diameter. S2, S3, S4 are after Shimizu (1960); Z, LR, CC and MAG 5 are after Tivey & Johnson (1981, 1983);all other points from the present study.

anticipated on theoretical grounds and is seen in Tivey & Johnson's results but the precise size range over which it occurs is better defined by the new data.

VM acquisition apparently remains at a low, essentially constant level between 0.1 and 2 μm, then increases two-fold in going from 2 to 14 μm. If this peak is real, it presumably is related to size-dependent domain structures in this range.

The peak may not be real, however. The 10 and 14 μm samples have high values of S_a but lower, more normal values of S_d, close to the average S_d value over the 3 decades 0.1–100 μm (indicated by the dashed line in Fig. 1). Higher-than-average values of S_d occur instead in 4 and 6 μm grains.

An unmistakable feature of the decay data is the high value of S_d for the 0.037 μm (small SD) grains. Mullins & Tite (1973) determined S_d, both directly and from quadrature AC susceptibility, for a sample of $\leqslant 0.03$ μm magnetite. Their results, in arbitrary units, convert (using measured susceptibilities) to a value $S_d \approx (6.1 \pm 0.2) \times 10^{-2}$emu cm^{-3}, about twice the value measured for the 0.037 μm sample. In view of the pronounced size dependence of S_a and S_d in this range – approximately as $d^{-1.3}$ – and the fact that the smallest grains in either sample are probably responsible for most of the magnetic viscosity, the agreement is not unreasonable.

TIME DEPENDENCE OF VM AND VRM

VM acquisition and VRM decay measured at temperatures from 9 to 500°C are depicted in Figs 2, 3 and 4. The time-scale is logarithmic in each plot. If VM and VRM changed in proportion to log *t*, the data would fall on straight lines. Instead the acquisition and decay characteristics are curved. VM acquisition and VRM decay both accelerate with time compared to a log *t* function, or equivalently, the instantaneous viscosity coefficients $S_a = \partial J/\partial \log t$, $S_d = |\partial J_r/\partial \log t|$ increase with time. Non-linearity is most pronounced at high temperatures.

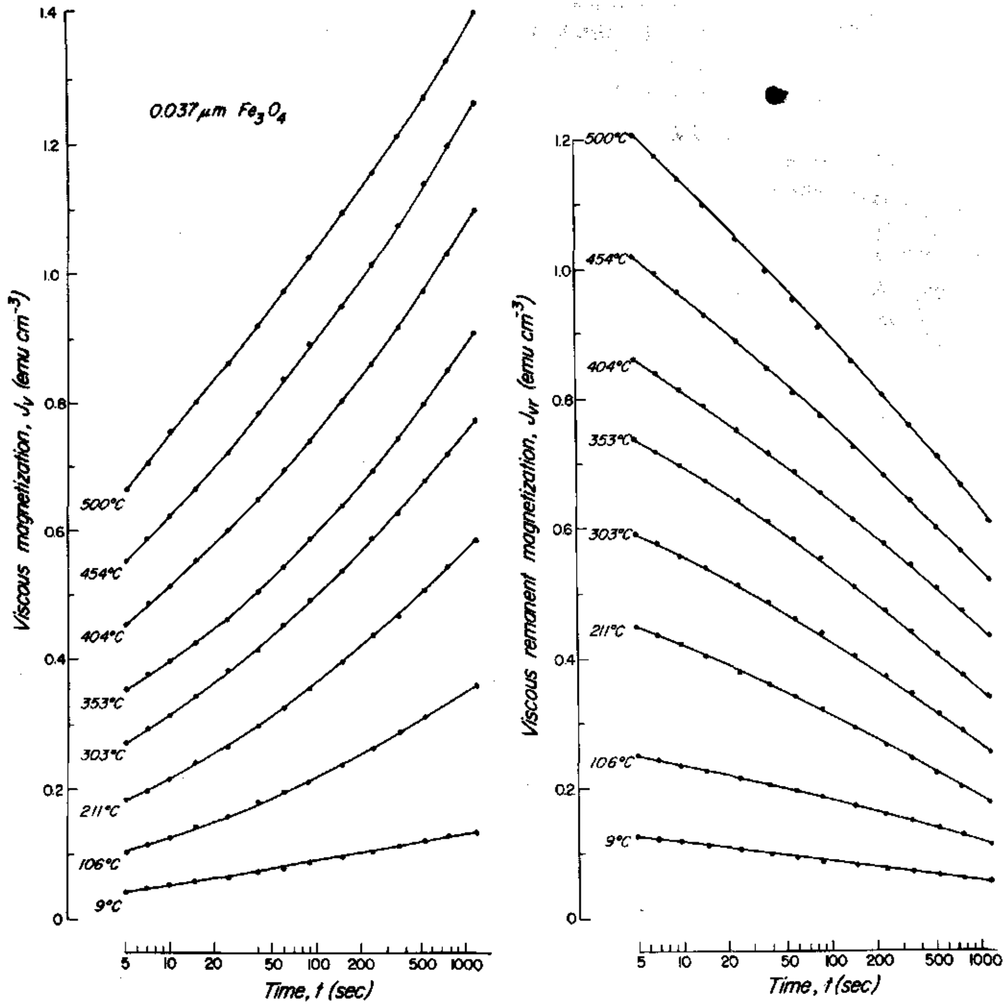

Figure 2. Acquisition of VM and decay of VRM at various temperatures for the 0.037 μm sample. The zero of the ordinate is arbitrary and the curves have been separated arbitrary amounts for convenience in drawing. Approximately log t behaviour is found at 9 and 500°C but at intermediate temperatures viscosity coefficients or slopes $|\partial J/\partial \log t|$ increase with time.

The curvature is not a result of non-linear response of the magnetometer or recording system. The degree of curvature depends on sample temperature and is therefore a sample property, since the magnetometer and electronics remained at room temperature during all runs. Systematic temperature increases during a run cannot explain the observations either. S_a or S_d at the end of one run is usually $\geqslant S_a$ or S_d at the beginning of the next run, at a temperature 50 or 100°C higher. Temperature drift of 1–2°C during a run could not always be avoided, but large systematic temperature changes certainly did not occur.

Tivey & Johnson (1981, 1983) observed accelerating VM acquisition like that illustrated in Figs 2–4, although their VRM decay curves are nearly linear in log t. Their measurements began and ended at longer elapsed times than in the present experiments. Plessard (1971) produced strikingly different viscosity coefficients, in a single tephrite sample at a single temperature, by varying the zero-field wait time following thermal demagnetization and preceding VM acquisition. As S_a or S_d increased, so did the curvature of the acquisition or

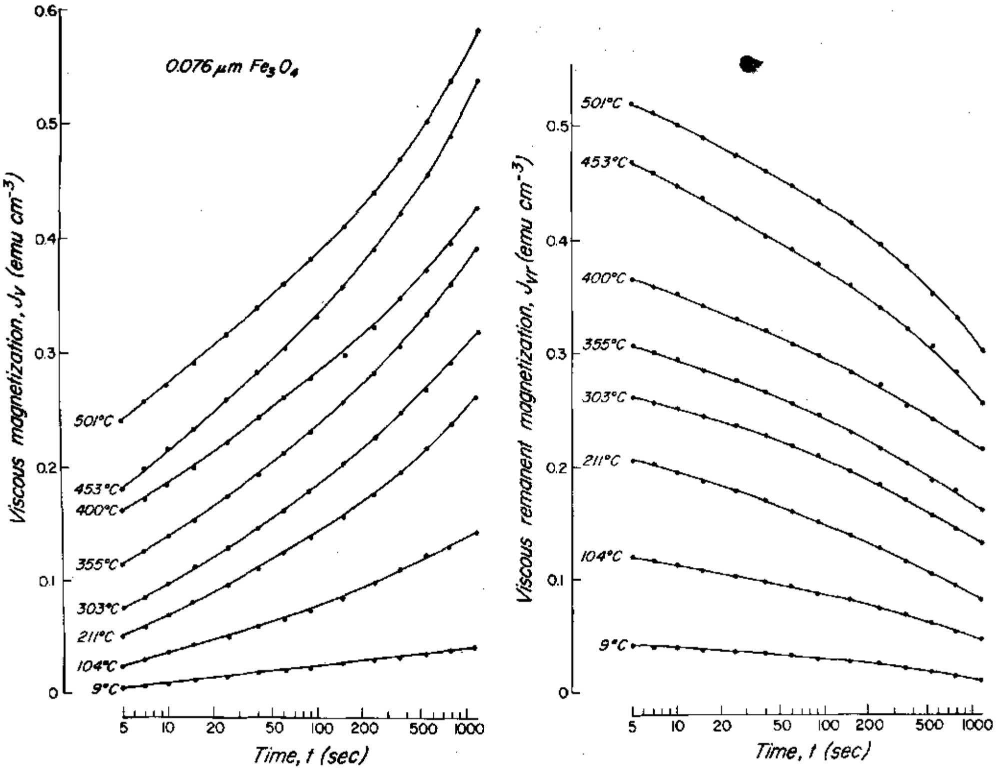

Figure 3. VM acquisition and VRM decay at various temperatures for the 0.076 μm sample. Note the expanded scale of the ordinate compared to Fig. 2.

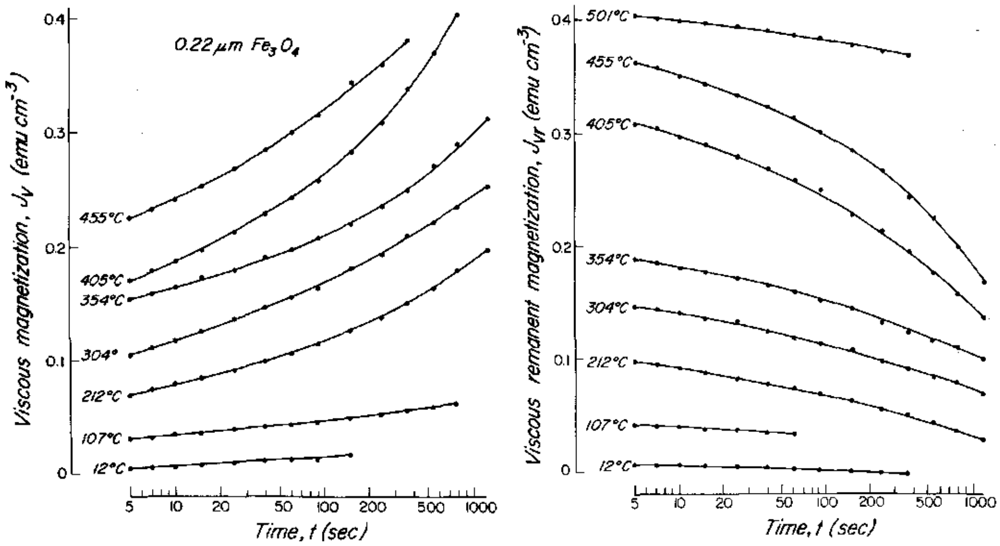

Figure 4. VM acquisition and VRM decay at various temperatures for the 0.22 μm sample. The scale of the ordinate is the same as in Fig. 3.

decay characteristics. Since the same sample was used in each experiment, the effect seems to be fundamental.

Tradition to the contrary, it may be that the log *t* dependence of magnetic viscosity is not fundamental. The non-log *t* part of VM or VRM is, however, not very prominent unless the viscous changes are intrinsically large or are amplified by measuring over extended times or at high temperatures. Lowrie & Kent (1978), Dunlop & Hale (1977), Dunlop & West (1969), and Ozdemir & Banerjee (1981) all recorded increases in S_a or S_d with time in intrinsically rather viscous samples, particularly at above-ambient temperatures or long times, but essentially log *t* behaviour in only slightly viscous samples.

To obtain quantitative measures of the log *t* and non-log *t* parts of VM and VRM, the data of Figs 2–4 were fitted to a quadratic function

$$J, J_r = a \log t + b\,(\log t)^2 \qquad (7)$$

A quadratic form was suggested by the aspect of the VM and VRM curves and by basic theory, e.g. equations (5) and (6). A least-squares quadratic spline program on a HP programmable calculator/plotter was used for curve fits. The solid curves in Figs 2–4 are examples of the results. The fits are excellent and the two parameters *a* and *b* adequately describe the physical phenomenon.

The data and curve fits can also be presented in the form or linear plots of viscosity coefficient S_a or S_d versus log *t*. Fig. 5 is an example of such a plot for the 0,037 mkm sample. From (7), we have

$$|\partial (J, J_r)/\partial \log t| = S_{a,d} = a + 2b \log t. \qquad (8)$$

Thus a, which is a measure of log t behaviour, is the intercept or 'initial' value (t = 5 s in these runs) of S and b, which is a measure of curvature or non-log t behaviour, is proportional to the slope $\partial S/\partial \log t$.

It is evident from Fig. 5 that *a* and *b* are correlated, just as they were observed to be in the work of Plessard (1971). High values of slope *b* tend to accompany higher values of intercept *a*. The correlation is more evident in acquisition characteristics, *b* values (i.e. curvatures) being much smaller in decay.

TEMPERATURE DEPENDENCE OF VM AND VRM

Figs 2-4 illustrate the considerable enhancement of magnetic viscosity at higher-than-ambient temperatures. Fig. 6 gives the explicit temperature dependences of the average viscosity coefficients S_a or S_d for these samples. The variation with temperature of initial susceptibility $_0$, determined by dividing IM at the beginning of each run by the applied field H_0 = 10 Oe, is also shown for comparison.

Xo increases in a regular fashion with increasing temperature, approaching a Hopkinson peak by 500°C. (The apparent decrease in Xo at high temperature in the 0.22mkmsample is an irreversible effect, probably resulting from oxidation as discussed earlier.) S_a or S_d on the other hand, do not increase in a regular fashion, contrary to the predictions of all simple theories. There seems to be a threshold near room temperature below which magnetic viscosity is unimportant, rapid increases between 100 and 200°C and again between 350 and 450°C with a 'plateau' of low $S/$ between, and a tendency to saturation by 500°C.

High temperature saturation is easily explained. Thermally activated viscous effects should slacken and eventually cease once all relaxation times *t* become t_{exp}, the duration of an experiment, here 20 min. This takes place in the peak blocking temperature range,

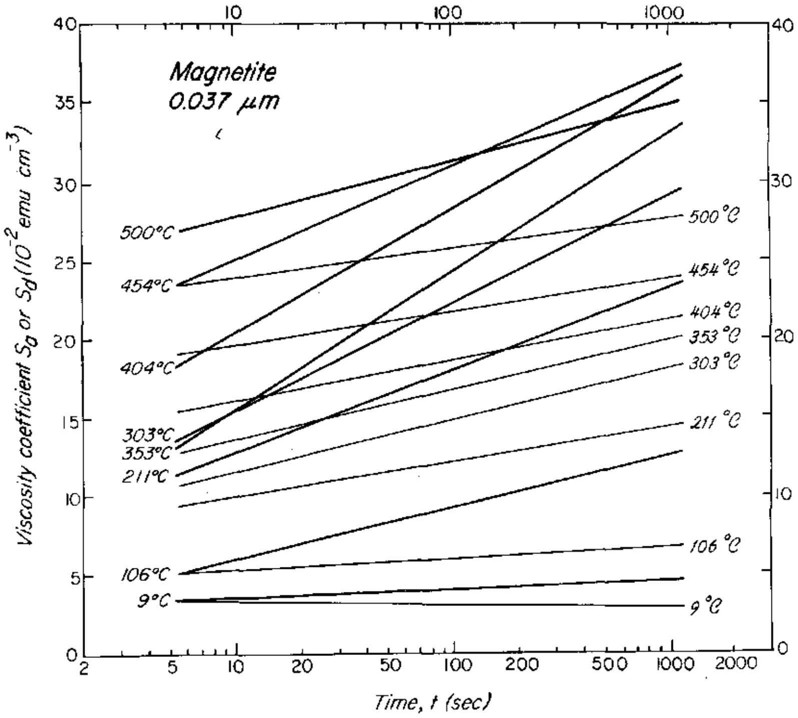

Figure 5. A plot of viscosity coefficients as a function of time for the 0.037 Mm sample that embodies the same information as Fig. 2. Individual data points have been omitted to avoid confusion but the fits are of similar quality to those in Fig. 2. Heavy lines, S_a; light lines and script figures, S_d. The linear dependence of S_a or S_d on log *t* demonstrates a quadratic dependence of VM amd VRM on log *t* in Fig. 2.

which is 545-570°C for these samples (Dunlop 1973c; Dunlop & Bina 1977), However, under these same conditions r $\lesssim t_{\mathrm{exp}}$, the susceptibility resulting from reversible domain rotations or wall displacements will rise toward a superparamagnetic peak (Dunlop 1974).

The plateauing tendency around 300°C is seen in all four submicron samples but has no obvious explanation. Once must be cautious in ascribing undue significance to minor trends in viscosity coefficients but these trends do seem to occur consistently. There were no systematic variations in factors that could have modified viscosity coefficients, such as zero-field wait time between runs, following thermal or AF demagnetization (Plessard 1971; Tivey & Johnson 1981, 1983) or undemagnetized residual VRM from previous runs (Lowrie & Kent 1978).

The 'curvature coefficient' *b* also increases with temperature. However, its temperature variation (not plotted) is rather different from that of either *S* or Xo, reaching a peak around 350°C and decreasing at higher temperatures. This variation is quite apparent in the acquisition characteristics of Fig. 5. The log *t* and non-log *t* parts of VRM and VM therefore exhibit different temperature dependences, the former having a low rate of increase *dS/dT* in about the same temperature range that the latter peaks. The nature of magnetic viscosity seems to change with temperature, or more precisely, if one assumes for convenience independent physical origins for log *t* and non-log *t* behaviour, the partitioning between these two sources of magnetic viscosity is different at different temperatures.

me $\lesssim t_{\mathrm{exp}}$, the duration
temperature range,

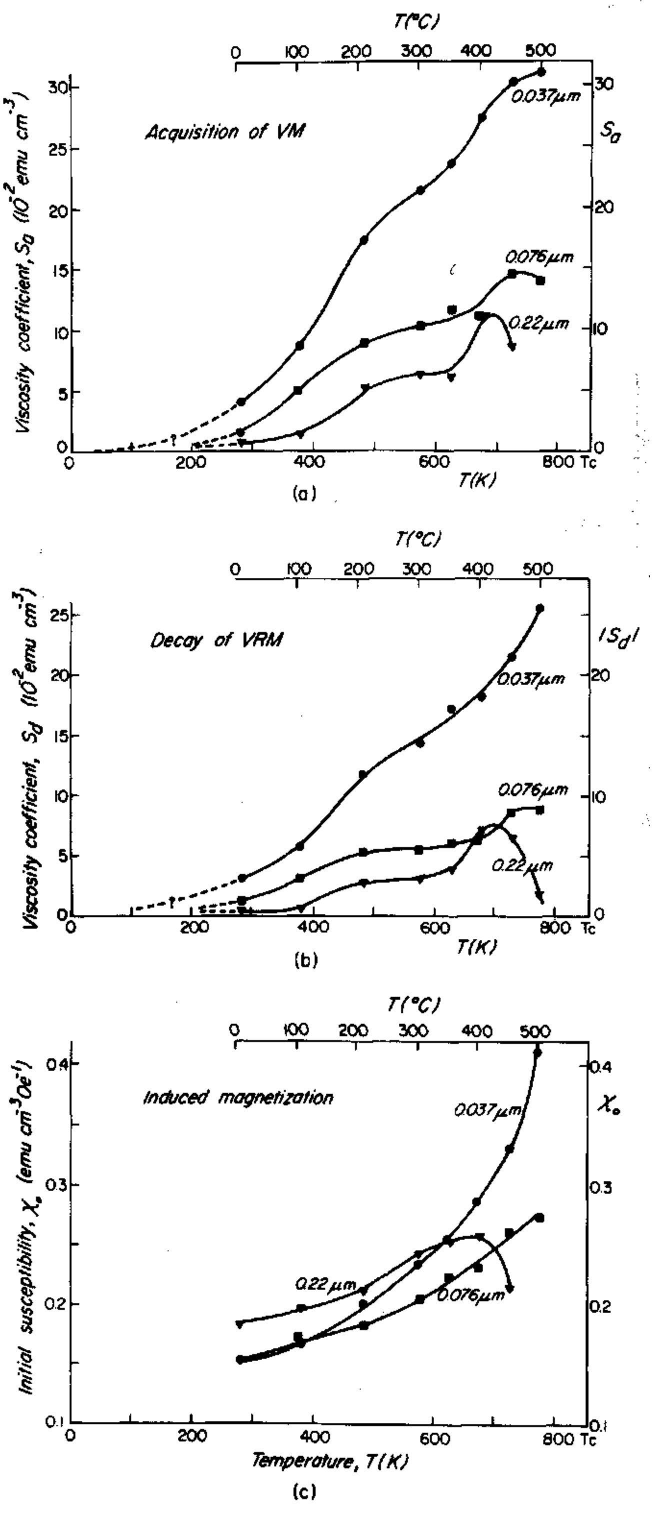

Figure 6. Temperature dependence of (a) average VM acquisition coefficient S_a, (b) average VRM decay coefficient S_d, (c) initial (10 Oe) susceptibility χ_0 for the submicron samples of Figs 2–4.

The evidence from the literature is far from clear-cut. Shimizu's (1960) very detailed measurements on rather coarse crushed magnetites (average grain sizes * 270, 100 and 2 mkm for samples S1, S2 and S3) supported Street & Woolley's theory. Between -100 and 400-500°C, S_a was proportional to T. A Hopkinson-type peak occurred just below the Curie point, presumably reflecting the $J_s^{-2}(T)$ factor in the Street & Woolley theory. There were no consistent intermediate-temperature trends. Shashkanov & Metallova (1970), on the other hand, found both low-temperature (100–200°C) and high-temperature viscosity peaks. Their results perhaps express an extreme form of the trends seen in Fig. 6.

Relations between acquisition and decay characteristics

According to Neel (1950), the ratio of S_a for VM acquisition to S_d for VRM decay is diagnostic of domain structure. For domain wall displacements in the weak-field Rayleigh region, S_a should equal 2 S_d (Neel 1950), but for rotations of SD moments, $S_a = S_d$ (Neel 1949).

Experimental values of the ratio S_a/S_d are relatively insensitive to temperature in individual submicron-size samples (Fig. 7a). As predicted, the ratios are almost always between 1 and 2 and they vary from one sample to another. However, there is not the monotonic dependence on grain size that one would expect for a parameter sensitive to domain structure.

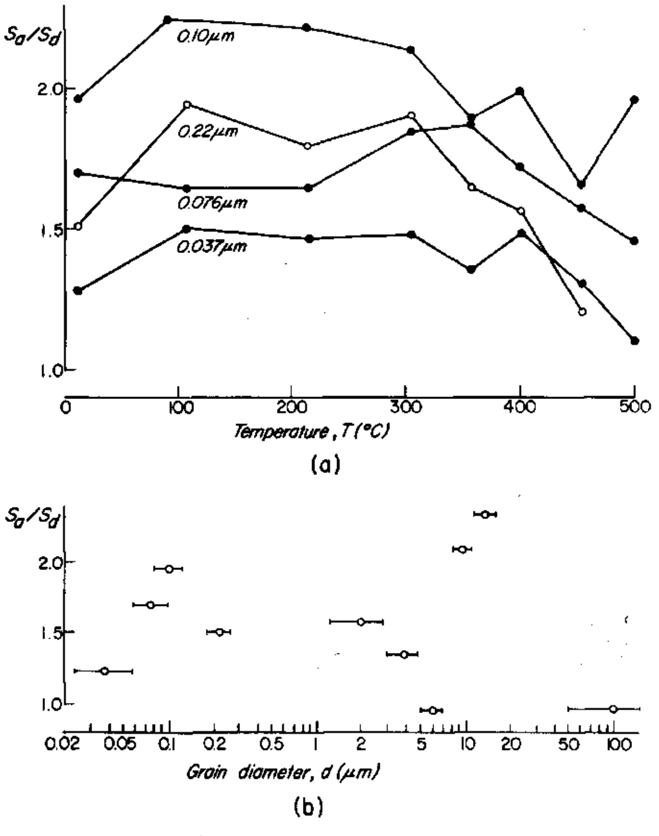

Figure 7. The variation of the ratio S_a/S_d of VM acquisition and VRM decay coefficients as a function of (a) temperature and (b) grain size.

Néel's predictions about S_a and S_d are really predictions about the relative magnitudes of VM (J) and VRM (J_r). It was implicitly assumed that the same range of relaxation times is involved in decay as in acquisition, i.e. that VRM acquired in a 20 min exposure to H_0 will completely decay during 20 min in zero field. This is never the case, even in principle, and in the present experiments as much as 10–15 per cent of the initial VRM remained at the end of some 20 min decay runs.

There are two reasons for this behaviour. First,$(H_0) <$ (0) because even a small field H_0 reduces the energy barrrier to be overcome by thermal energy. The reduction is not necessarily negligible for a field as weak as 10 Oe, particularly for MD grains (see Dunlop 1973a, pp. 872-873). The second reason is that thermal activation is a random process. Consider an ensemble of identical SD moments or pinned domain walls, each characterized by same tau. After exposure for a time $t = tau$ to H_0[assumed to be so weak that$(H_0)^{\iota}=$ (0)], 1 — ($1/e$) or 63 per cent of the ensemble will have been thermally activated. Of these activated grains or walls, after a time $t = tau$ in zero field, 63 per cent will have returned to their original state, leaving 37 per cent in an activated state. Of course, most of the ensembles activated in the present experiments have tau $\ll$ 20 min but the random reactivation principle remains valid.

As Fig. 5 makes clear, the ratio of b during VM acquisition to b during VRM decay is considerably greater than the ratio $/S_d$. In other words, deviations from log t behaviour are much more prominent during acquisition than during decay (*cf.* Tivey & Johnson 1981, figs 2 and 3).

THERMAL DEMAGNETIZATION OF UNDECAYED VRM

In order to thermally demagnetize undecayed VRM after a run at temperature T_1, it should have been sufficient to heat the sample 50 or 100°C to the temperature $_2$ of the next run and maintain this higher temperature for a relatively short time. Taking the average tau$_1$ of

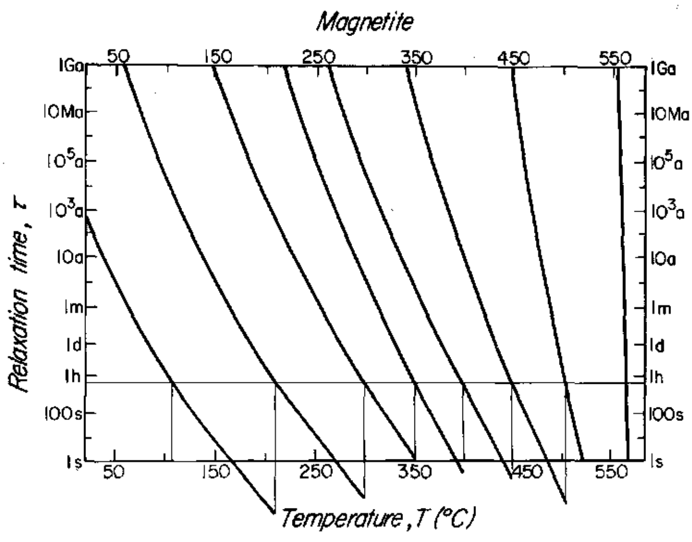

Figure 8. Blocking contours joining equivalent (τ, T) pairs for particular SD grain ensembles, calculated following Pullaiah *et al.* (1975) or equation (9) of the text. The contours shown are appropriate to ensembles carrying VRM with $\tau_1 \approx 20$ min (light horizontal line) at temperatures T_1 of 105, 210, 300, 350, 400, 450 and 500°C (light vertical lines). The relaxation time τ_2 of each ensemble at the next highest temperature T_2 is $\lesssim 1$ s. Each VRM should therefore be destroyed by zero-field heating from T_1 to T_2.

undecayed VRM at T_1 to be 20 min (an overestimate), Fig. 8 predicts that the corresponding value of relaxation time$_2$ at T_2 should be < 1 s for all (T_1, T_2) used in successive runs in the present study.

Thus the undemagnetized residual VRM at T_2 after timetau$_2$~1 s should have been zero for all entries in Table 2. The samples were actually annealed at least 30 min at each higher temperature to ensure that all parts of the sample equilibrated at T_2. In practically every case, the residual VRM at T_2 was not ≈ 0 but 8-35 per cent of the undecayed VRM at T_1.

The curves of Fig. 8 are based on the SD theory of Néel (1949), following calculations by Pullaiah *et al.* (1975). The sharp curves shown are averages only, since relaxation is a gradual phenomenon (see Dunlop 1981, fig. 17 for curves of 5 and 95 per cent relaxation). However, randomness of reactivation discussed in the last section is an inadequate explanation of the large residuals in Table 2. Viscous decay of VRM in an SD ensemble with a single value of τ covers a broad time range but thermal decay occurs over a narrow range of unblocking temperatures (10–20°C at most).

Of course SD theory may be inappropriate. Three of the four samples listed in Table 1 have mean grain sizes larger than nominal critical SD size (0.05 μm) and all four have J_{rs}/J_s values well below the SD value of 0.5. Now as discussed in the theory section, SD and MD formulations of thermal activation phenomena are very similar. However, there is one significant difference. A domain wall pinned at temperature T_B in field $\mathbf{H}_0$ begins to move towards a demagnetized state upon heating in zero field to an unblocking temperature T_{UB}, but a series of further Barkhausen jumps at higher temperatures is required for complete demagnetization (V. P. Shcherbakov 1982, private communication). Although there is as yet no formal theory of the T_{UB} spectrum associated with a single T_B, the effect is well documented experimentally (Bolshakov & Shcherbakova 1979). Only the lowest T_{UB} is related to T_B in the manner of Fig. 8 and substantial residuals are to be expected at $T_2 = (T_{UB})_{min}$

Table 2. Undemagnetized residuals in thermal demagnetization (at T_2 for a minimum time$_2$) of VRM acquired at$_1$ in time tau$_1$.

Sample grain size (Mm)	T_1 (°C)	tau$_1$ (s)	T_2 (°C)	tau$_2$ (s)	Residual VRM (per cent) Predicted	Residual VRM (per cent) Measured
0.037	106	1200	211	1	0	10.2
	211	1200	303	1	0	11.4
	303	1200	353	1	0	33.3
	353	1200	404	1	0	18.2
	404	1200	454	1	0	18.5
	454	1200	500	1	0	10.3
0.076	104	1200	211	1	0	3.3
	303	1200	355	1	0	29.0
	355	1200	400	1	0	22.4
	400	1200	453	1	0	26.3
	453	1200	501	1	0	31.8
0.10	352	1200	399	1	0	30.3
	399	1200	449	1	0	32.1
	449	1200	500	1	0	33.3
	303	1200	500	1	0	0
0.22	212	1200	303	1	0	7.5
	354	1200	405	1	0	35.2
	304	1200	455	1	0	11.1
	455	1200	501	1	0	10.5

Discussion

The grain size dependence of VRM is difficult to characterize with certainty. From Fig. 1 it is clear that both SD and MD grains are significantly viscous. Yet the room-temperature VRM acquired by the most viscous sample, containing SD grains averaging 0.037 μm in size, during 20 min exposure to a field *(21/2* decades of time) is only 1-2 per cent of TRM acquired in the same field (Dunlop 1973c). VRM at ordinary temperatures tends to be inefficient, involving a small fraction of the relaxation time spectrum (log τ). (At high temperatures, of course, larger fractions of *f*(log*tau*) are activated and VRM increases.) Among SD grains, only these near superparamagnetic size have tau ~ *t* and react to fields applied for time *t.*

Among MD grains, grain size *per se* is not the controlling factor, since activation of an entire domain wall in a > 1 μm grain is energetically most unlikely (Dunlop 1973a, pp. 887-888). Thermal activation of metastable SD grains of this size (Halgedahl & Fuller 1980) is more unfavourable still. If segments of bowed walls firmly pinned at several points are the units activated, as seems probable (Craik & McIntyre 1967; Gaunt & Mylvaganam 1979), pinning strength and distance between pins must be the factors that control VRM. These factors depend more on impurities, state of stress and previous history of a sample than on grain size.

Tivey & Johnson (1981) found a four-fold increase in $_a$ over the size range ~ 0.2 to ~ 0.02 mkm in magnetite. Their magnetites were of varied provenance (data for one of their samples, containing grains with 8:1 elongations and resulting high coercive forces, are omitted from Fig. 1). The submicron monocrystal cubes used in the present study, narrowly sized and all prepared in similar fashion, show the same four-fold increase in S_a but over a narrower range, from 0.1 to 0.04 mkm.

There is nevertheless some doubt whether either experimental trend truly reveals the intrinsic size dependence of SD magnetic viscosity. The 0.037 μm sample contains crystals as small as 0.02mkmand as large as 0.07mkm.Tivey & Johnson's sample Z has a smaller average crystal size (0.016 mkm), but there is doubtless a considerable spread of sizes, although no figures are given. These two samples have similar $_a$ values, which likely characterize the larger crystals in sample Z and the smaller crystals in the 0.037 mkm sample. Since the size spread of even the best experimental samples exceeds the probable size range for viscous behaviour, it is impossible to draw more definite conclusions.

The larger submicron magnetites have size ranges that nominally exclude 0.02–0.04 μm, but their capacity to acquire VRM remains significant. Is their viscosity due to a trace fraction of very small crystals, which tend to be masked by larger crystals or to remain unresolved in electron micrographs? Or are these grains inhomogeneously magnetized, containing a vestigial domain wall? Since theoretically two-domain grains possess independent SD-like and MD-like modes of magnetization change (Dunlop 1977, 1981), the VRM, if it is intrinsic to this size range, may not be simple.

Relatively large MD grains also acquire appreciable VRM. The steady increase in $_a$ between 2 and 14 μm (Fig. 1) may result from weakening wall pinning, as indicated by decreasing coercive forces (Table 1). However, ~ 100 mkm magnetites, with the lowest H values of all, acquire less VRM than smaller grains. Of course, wall pinning is not the only governing factor. The longer ball-milling needed to produce smaller grains may produce more pinning centres and smaller, more readily activated wall segments. The competing effects of activation volume and pinning strength may be at the root of the contradictory size dependence of viscosity coefficients in 2-20 mkm magnetites reported by Zhilaeva & Minibaev (1965). Very careful design of future experiments will be required before we can move out of the realm of speculation, however.

The time-honoured $\log t$ dependence of viscous magnetization changes (e.g. Thellier 1938) is not supported by the results of Figs 2–4. Instead a quadratic in $\log t$, $J = a \log t + b (\log t)^2$, describes the acquisition of VM and loss of VRM over the time-scale 5–1200 s. The ratio b/a increases with increasing temperature. VM acquisition curves of similar aspect have been measured by Tivey & Johnson (1981) for both fine-grained and coarse-grained samples over more extended times (250–6×10^4 s). Shimizu (1960), on the other hand, found essentially $\log t$ behaviour at all temperatures for $\approx 2\,\mu$m magnetite but the experiments covered only 1½ decades of time (60–2000 s).

A quadratic law is not actually inconsistent with Néel's (1949) SD theory. Rather than a uniform distribution $f(\log \tau)$, which leads to a $\log t$ dependence, $f(\log \tau)$ must increase linearly with increasing $\log \tau$: $f(\log \tau) = \alpha + \beta \log \tau$. In fact, Fig. 5 is a more or less direct mapping of $f(\log \tau)$ at increasing values of τ, as revealed at higher and higher temperatures.

Néel's (1950) MD theory and Walton's (1980) SD theory provide more natural explanations, without the need for a special distribution of τ. For the Néel theory, expanding equation (5),

$$J_{\mathrm{vrm}}(t) = H_0 p S_{\mathrm{v}} \log t + \tfrac{1}{2} p S_{\mathrm{v}}^2 (\log t)^2,$$

so that $b/a = S_{\mathrm{v}}/2H_0$. For the Walton theory, from equation (6),

$$J_{\mathrm{vrm}}(t, T) \cong CH_0 [T/K(T)]^{2+r} [(\log f_0)^{3+r} + (3+r)(\log f_0)^{2+r} \log t$$
$$+ \tfrac{1}{2}(3+r)(2+r)(\log f_0)^{1+r} (\log t)^2]$$

since $\log f_0 \approx 23 \gg \log t$ for $t \lesssim 10^3$ s. In this theory, $b/a = (2+r)/2 \log f_0$ and the T and t dependences of VRM are coupled.

The Walton theory has an obvious drawback in that r is a constant related to the grain size distribution and so b/a should be temperature independent, contrary to observation. If $r = 0$ (uniform distribution of V or of $\log \tau$), $b/a = 0.043$ and if $r = -1$ (close to a log normal distribution at large V), $b/a = 0.022$. The experimental temperature dependence of S_{a} is not simple (Fig. 6) but a T^2 law, corresponding to $r = 0$, would be a reasonable representation at low temperatures.

The Néel MD theory is more promising in that $S_{\mathrm{v}} \propto T^{1/2}$ at temperatures far from the Curie point, so that b/a should increase with temperature, as is observed. The magnitude of b/a is difficult to estimate, since p is required to calculate ${}_{\mathrm{v}}$ and the second Rayleigh coefficient was not determined in these experiments. Similarly the temperature dependence of S_{a} and S_{d} cannot be tested unless $p\,(T)$ is determined.

It has been assumed throughout this paper that magnetic viscosity can be ascribed wholly to thermal fluctuations. A competing effect could be diffusion after-effect (Tropin & Vlasov 1966; Moskowitz & Banerjee 1981), due to diffusion of vacancies that help pin domain walls. The superposition of two quite different physical mechanisms could well produce a complex temperature dependence of magnetic viscosity. Since the activation energy for vacancy diffusion in magnetite is quite low, diffusion after-effect might contribute to enhanced short-term VRM in the 20-200°C range, but fluctuation after-effect must be of paramount importance at high temperatures and long times.

The acquisition of VM proceeds more rapidly and deviates more from a $\log t$ law than the corresponding decay of VRM. Values of S_a/S_{d} vary irregularly with grain size (Fig. 7) and lend no particular support to either SD or MD theory.

Néel's (1949) SD theory does not adequately describe thermal demagnetization of VRM. The relation

$$\frac{T_1}{K(T_1)} \log f_0 t_1 = \frac{T_2}{K(T_2)} \log f_0 t_2 \tag{9}$$

(Pullaiah *et al.* 1975), graphed in Fig. 8, predicts that VRM blocked in time t_1 at T_1 should be erased by zero-field heating to T_2 and annealing for time τ_2. In practice, at the predicted temperature T_2 even for times well in excess of τ_2, up to 35 per cent of the VRM survives (Table 2).

One possible explanation is that (9) is wrong. Walton's (1980) SD theory leads to the relation

$$\left[\frac{T_1}{K(T_1)}\right]^{2+r} (\log f_0 t_1)^{3+r} = \left[\frac{T_2}{K(T_2)}\right]^{2+r} (\log f_0 t_2)^{3+r} \tag{10}$$

which predicts higher values of T_2 than (9). Alternatively if VRM is essentially an MD, wall activation phenomenon in 0.04– 0.22 mkm grains, there may be no single T_2 but a broad thermal demagnetization tail extending from approximately the value given by (9) to very high temperatures (Bolshakov & Shcherbakova (1979).

On the whole, a single-valued relation between T_2 and T_1 seems more probable, since prolonging the anneal at T_2 to ~ 60 min or raising the temperature 20–30°C was always sufficient to erase the residuals in preparation for the next run.

Conclusions and implications for palaeomagnetism

The results reported in this paper provide some new insights into the nature of viscous magnetization and also reveal some areas where present understanding of the underlying mechanisms is weak or faulty. The practical implications for palaeomagnetism can be summarized as follows.

(1) VRM is a property of magnetites (and undoubtedly other minerals too) of all sizes. Nearly superparamagnetic SD grains and 5–15 mkm MD grains are exceptionally viscous but VRM at a lower level is a contaminant of other size ranges as well. VRM may even overshadow stable NRM in rocks containing exclusively ultrafine grains, such as lunar soils and soil breccias (Gose *et al.* 1972), or relatively coarse MD grains, such as some submarine massive basalts (Lowrie & Kent 1976, 1978; Deutsch & Pätzold 1976) and many felsic plutonic rocks.

(2) VRM is produced at an accelerated rate if the temperature is even slightly elevated. Viscosity coefficients increase more as T^2 than as T just above room temperature. Basalts erupted at mid-ocean ridges during the Brunhes epoch may, for this reason, have acquired more VRM than their room-temperature viscosity coefficients would suggest, while basalts erupted in previous epochs may preserve a residual fraction of their earliest VRM, normal or reversed.

(3) Since the time-scale for decay is longer than the time-scale for acquisition of VRM, it cannot in any case be assumed that VRM produced during one polarity epoch will completely vanish during the following epoch.

(4) VM acquisition accelerates with time compared to a log t law. This non-log t behaviour was found in all grain sizes examined, although it was most obvious in very viscous samples and at elevated temperatures. Extrapolations that use a laboratory determination of viscosity coefficient and an assumed log t law are likely to underestimate, perhaps even grossly underestimate, the VRM acquired over times of geological length. In extreme cases, S_a doubles or triples over 2-3 decades of time (see Fig. 5 or Tivey & Johnson 1981, figs 2 and 3).

Of course there is no guarantee that S_a continues to increase at very long times, but laboratory determinations of S_a are unlikely to be representative. The safest method of extrapolation is to activate rs of geological length directly by measuring VM or VRM at above-ambient temperatures (Dunlop & Hale 1977).

(5) Thermal demagnetization is somewhat less efficient than predicted in erasing VRM (Table 2) but nevertheless much more efficient than AF demagnetization. On the few occasions when samples were cooled and their high-temperature VRMs AF cleaned, fields of at least 1000 Oe were required, whereas a heating of 50–100°C above the acquisition temperature would have achieved the same end.

(6) Equation (9) underestimates the reheating temperature needed for short-term thermal demagnetization of a long-term VRM or VPTRM acquired at lower temperatures. The discrepancy is exactly similar to the one reported by Pullaiah *et al.* (1975) in their original paper. The implications are serious because equation (9), its graphical equivalent Fig. 8, or other similar formulations (Dodson & McClelland-Brown 1980) are widely used (e.g. Schwarz 1977; Van der Voo, Henry & Pollack 1978; Buchan & Schwarz 1981) to calibrate the temperature of thermoviscous overprinting of NRM in metamorphic rocks by comparison with laboratory thermal demagnetization behaviour.

More serious still, if T_{UB} is not a single-valued function of T_{B}, the standard test for thermal or thermoviscous overprinting, namely non-overlapping T_{UB} spectra of primary and overprint VRMs, cannot be expected to hold in rocks whose remanence is of essentially MD character. Although this point is outside the scope of the present paper, careful VRM experiments (which can isolate a narrower range of *tau* than partial TRM experiments) are probably the best means of clarifying this important question.

Acknowledgments

I am grateful to Dr Claude Plessard for allowing me to use his equipment and for his interest in the experiments. Maxime LeGoff cheerfully offered his expert assistance at all stages of the work. The cooperation offered by these and other members of the St-Maur laboratory contributed greatly to the success of the project. Dr Valera Shcherbakov brought to my attention important work on diffusion after-effect and distributed unblocking temperatures of MD grains. This research was supported by the Natural Sciences and Engineering Research Council of Canada through a travel grant during my stay in France and research grant A7709 during the writing up.

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