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# Magnetic viscosity, diffusion after-effect, and disaccommodation in natural and synthetic samples

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**Summary.** The magnetic viscosity of some oceanic basalts from IPOD site 417D is investigated. The samples are fine-grained pillow basalts and coarse-grained massive flows. Experiments on the viscous behaviour of these samples include (1) acquisition and decay of weak-field viscous remanent magnetization (VRM) and (2) the influence on VRM produced by the zero field storage time following demagnetization by an alternating magnetic field. In all cases the aquisition of VRM is found to be more rapid than its subsequent decay. The time dependence of the intensity of VRM is observed to be significantly non-logarithmic during acquisition, but less so during decay. The length of time a sample remains in zero field following demagnetization produces a decrease in the aquisition and decay coefficients of VRM. These results cannot be reconciled entirely with predictions based on existing models of magnetic viscosity due to thermal fluctuations. Available evidence also suggest that the effect on VRM produced by (1) zero field storage following demagnetization or produced by (2) different initial states of magnetization, is attributed to susceptibility disaccommodation. Unfortunately, the available data are insufficient fully to ascertain the importance of diffusion after-effect and disaccommodation to long-term viscous behaviour. It is suggested, however, that diffusional processes may contribute to VRM in young pillow basalts (<0.1Ma) near mid-ocean ridges where the oxidation of titanomagnetite produces an increase in the number of octahedral lattice site vacancies and the elevated temperature of the crust enhances the diffusive reordering of these vacancies and ferrous ions. In order to gain some additional insights into VRM and attempt to explain some of these observations, theoretical and experimental results pertaining to diffusion after-effect and disaccommodation in magnetic materials are reviewed.

## 1 Introduction

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Time-dependent changes in the magnetization of ferromagnetic materials are commonly referred to as magnetic viscosity or magnetic after-effects, the study of which has had a long history of both experimental and theoretical research. Pioneering experiments by Rayleigh

(1887) and Ewing (1889) followed by the important theoretical studies by Preisach (1935), Richter (1937), Neel (1949, 1950, 1951) and Street & Woolley (1949), set the stage for the continuing investigations of magnetic viscosity. The fact that research continues after l00 yr serves to demonstrate that although our basic understanding of the viscous behaviour of magnetic materials is sound, a comprehensive theory is still lacking.

Various mechanisms contribute to the viscous behaviour of magnetic substances and include the following: (1) thermal fluctuations; (2) diffusion after-effect associated with the diffusion of ferrous ions and vacancies; (3) time-dependent changes associated with chemical alteration; and (4) eddy currents. For rock magnetism, thermal fluctuations have traditionally been assumed as the primary source for viscous changes in magnetic minerals. I wish to point out however, that diffusional after-effects, an important source for viscous behaviour in metals, alloys, and commercial ferrites (magnetic oxides), can also contribute to time-dependent changes in magnetite and titanomagnetite. There has been much research into both the commercial and scientific aspects of diffusion after-effect in a wide variety of magnetic materials; nevertheless diffusion after-effect has received scant attention in the rock magnetism literature. A primary aim of this paper is to acquaint readers unfamiliar with diffusion after-effects, with the theoretical and experimental results of this research and to show how these results can perhaps provide additional insights into the viscous properties of rock magnetic materials. Time-dependent changes associated with chemical alteration, although important for rock magnetism, and eddy currents, which are not, will not be discussed further.

The thermal fluctuation model advanced by Neel (1949) and others helps to explain, and rather successfully, the remanent magnetization of single domain (SD) particles under the influence of (1) time, $t$ (known as viscous remanent magnetization or VRM), (2) temperature, $T$ (thermoremanent magnetization or TRM), and (3) magnetic field, $h$ (isothermal remanent magnetization or IRM). The thermal fluctuation model attributes VRM to the spontaneous reversal of an SD moment, or an irreversible displacement of a domain wall from a pinning site in a multi-domain (MD) grain. These fluctuations have characteristic relaxation times dependent on temperature, field, and activation volume (Street & Woolley 1949; N6el 1949; Gaunt & Mylvaganam 1979). In explaining the magnetic properties of MD particles, however, Noel's MD theory is not as successful. This is particularly apparent for VRM in MD particles (Dunlop 1973, 1983; Prevot 1981).

Thermal fluctuations, however, are not the only cause for time-dependent effects in magnetic materials. When a domain wall reaches an equilibrium position it is assumed to be in a local energy minimum. If, however, the subsequent diffusion of point defects, dislocations, impurity atoms, or electrons and their interactions with domain walls causes a rearrangement of the local spin orientations inside the wall, then the surface energy of the wall will change. As soon as the domain wall energy changes, it may no longer be in an energy minimum, and a displacement of the wall may result. This type of magnetic viscosity is called diffusion after-effect and was first observed in iron containing carbon or nitrogen impurities. Snoek (1947) was the first to relate this observation specifically to the diffusion of impurity ions. A rigorous theoretical treatment was given by N6el (1952).

Also associated with diffusion-related processes is a change in magnetic susceptibility with time following the application of a magnetic field. This phenomenon, call disaccommodation, is a well-known effect occurring in ferrites and is attributed, in part, to the diffusive reordering of vacancies and ferrous ions (Yanase 1962; Kronmiiller, Schiitzenauer & Waltz 1974).

A fundamental difference exists, however, between magnetic after-effect and disaccommodation. Magnetic after-effect (either due to thermal fluctuations or diffusion) causes a

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## 2 Theory

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change in the intensity of pre-existing magnetization, $J_{vrm}$. Disaccommodation (only due to diffusion), on the other hand, causes a change in the susceptibility, $x$, or in the ease of acquisition or change of magnetization. Disaccommodation will, therefore, lead to a stabilization of magnetization. Further, an important difference also exists between the after-effects due to thermal fluctuations and diffusion. The previous magnetic history of the sample strongly influences the diffusion after-effect. The longer the sample has been in zero field after a previous change in $h$, the smaller the increase in magnetization will be following a second application of a new $h$ (Brissonneau 1958; Sholpo 1966, 1967; Tivey & Johnson 1981,1984). An excellent demonstration of the importance of zero field storage time following demagnetization (or remagnetization) on VRM is given by Tivey & Johnson (1981, 1984) and this behaviour is referred to by them as the *dt* effect.

Thermal fluctuations, diffusion after-effects and disaccommodation are all thermally activated processes. Changes in magnetization due to the first occur spontaneously through direct random fluctuations, while in the latter two it is thermally activated diffusing species that produce the change in magnetization.

The purpose of this study is (1) to review briefly certain aspects of magnetic viscosity and disaccommodation and (2) to present new experimental data on these topics. Recent experimental investigations (Prevot 1981; Dunlop 1983; Tivey & Johnson 1981, 1984; Sporer 1984) have pointed out deficiencies in the thermal fluctuation model in explaining some aspects of VRM. Most notable are (1) the influence of the initial state of magnetization prior to VRM acquisition or decay, (2) the inequality of the acquisition ($S_a$) and decay ($S_d$) coefficients of VRM, and (3) the non-log $t$ behaviour of VRM. In practice, both (2) and (3) seem to be the rule rather than the exception. The new experimental work on VRM and disaccommodation is from a suite of oceanic basalts from site 417D (IPOD leg 52) and extends previous work described by Moskowitz & Banerjee (1981). To supplement these data, VRM results from synthetic titanomaghemites are also discussed. The data for the synthetic samples include primarily the results of Moskowitz & Banerjee (1981) and Ozdemir & Banerjee (1981).

A brief review of theoretical aspects of thermal fluctation and diffusion after-effects is given in Section 2. Sample description is briefly discussed in Section 3 and experimental procedures are outlined in Section 4. The experimental results are presented in Section 5. Section 6 discusses the results of this investigation and VRM results from the literature as they pertain to current thinking. Such conclusions as seem possible are stated in the final section.

## 2 Theory

### 2.1 THERMAL FLUCTUATION AFTER-EFFECT

Magnetic viscosity due to thermal fluctuations has been extensively reviewed bv Dunlop (1973). I will concentrate here on the predictions of the various VRM theories concerning the acquisitions ($S_a$) and decay ($S_d$) coefficients. The intensity of magnetization, to a first approximation, increases or decreases with the logarithm of time,

$$J_{vrm} \propto \log t. \tag{1}$$

The slope of this curve ($\partial J_{vrm}/\partial \log t$) is characteristic of the relaxation spectrum and is usually referred to as the viscosity coefficient ($S_a$ or $S_d$). This coefficient is also a function of $h$ and $T$ (Street & Woolley 1949; Shimizu 1960; Gose & Carnes 1973; Dunlop 1983). All VRM theories predict an equation similar to (1); the viscosity coefficient, however, may not be a constant (for a given $T$ and $h$) but may vary with time. This manifests itself in the

non-log f behaviour of $J_{vrm}$ observed by many authors (Dunlop & Hale 1977; Lowrie & Kent 1978; Tivey & Johnson 1981, 1984; Dunlop 1983). It should be obvious that as $t \to \infty$, equation (1) gives an unreasonable answer. One should expect, therefore, that as time increases $S_a$ (or Sj) should decrease. This is actually observed if the experimental time /a > maximum relaxation time $r_{max}$ or if the temperature is high enough to activate $\tau_{max}$ (Street & Woolley 1949; Dunlop & Hale 1977). Dunlop (1973, 1983) points out that log $t$ behaviour is predicted only by assuming some sort of uniform distribution of activation volumes, activation energies, or relaxation times. Tropin (1970) also demonstrates that by a judicious choice of distribution function any type of viscosity law can be obtained.

The point on which most VRM theories seem to agree is the symmetry between the acquisition and decay process, i.e. that $S_a = S_d$. Neel's (1950, 1955) viscosity field theory for MD grains is unique, however, because it makes the prediction that $S_a/S_d = 2$. According to Neel (1950, 1951) the effects of thermal fluctuations can be approximated by a fictitious fluctuating viscosity field, $H_v(t) = S_v(Q + \log t)$, where $S_v$ is the viscosity field coefficient and $Q$ is a constant (~ 50). Within the Rayleigh region ($h \ll h_c$, where $h_c$ is coercivity) the induced magnetization, $J_i$, is

$$J_i = \chi_i h + ph^2 \tag{2}$$

and Xi is the initial susceptibility andp is the second Rayleigh coefficient (e.g. Cullity 1972). The first and second terms of (2) represent the reversible and irreversible components of magnetization respectively. The viscosity field also produces an irreversible component (N6el 1950) and is assumed to be related to the irreversible susceptibility ($\chi_{irr}$) by

$$J_v = \chi_{irr} H_v(t) \tag{3}$$

where $J_v$ can be either the induced ($J_{vim}$) or remanent ($J_{vrm}$) viscous magnetization. In the Rayleigh region $\chi_{irr} = 2ph$ and therefore the total magnetization, given by the sum of the induced and viscous magnetizations, is

$$J = J_{vim} + J_i = \chi_i h + ph^2 + 2phS_v(Q + \log t_a) \tag{4}$$

and

$$\partial J_v / \partial \log t_a = S_{ai} = 2phS_v. \tag{5}$$

It is important to note that (4) and (5) refer only to an *induced* magnetization (indicated by the use of $S_{ai}$) and not to a remanent magnetization.

When $h = 0$, the isothermal remanent magnetization and irreversible susceptibility are, respectively, $J_{irm} = \frac{1}{2} ph^2$ and $\chi_{irr} = ph$. The total remanent intensity, in this case, is given by,

$$J = J_{irm} + J_{vrm} = \tfrac{1}{2} p\,[h + S_v(\log t_a - \log t_d))^2 \tag{6a}$$

and,

$$| \partial J_{vrm} / \partial \log t_a | = S_{ar} = phS_v + pS_v^2 (\log t_a - \log t_d), \tag{6b}$$

$$| \partial J_{vrm} / \partial \log t_d | = S_d = - phS_v - pS_v^2 (\log t_a - \log t_d) \tag{6c}$$

where $S_{ar}$ refers specifically to the case of a remanent magnetization. If $S_v/h \ll \log (t_a/t_d)$ then equations (5) and (6b, c) predict $S_{ai}/S_d = 2$ and $S_{ai}/S_{ar} = 2$. This prediction pertains only to the viscosity coefficients for VIM and VRM. If remanence measurements are made for both the acquisition and decay then equation (6a) is appropriate and predicts $S_{ar}/S_d = 1$ (e.g. Dunlop 1973).

**Table 1.** Theoretical predictions for VRM acquisition and decay coefficients.

| Theory | $S_{ar}/S_u$ | $S_{ai}/S_u$ | comment |
|---|---|---|---|
| Richter (1937) | 1 | 1 | SD and MD |
| Street & Woolley (1949) | 1 | 1 | SD and MD |
| Neel (1949) | 1 | 1 | SD |
| Neel (1950) | 1 | 2 | MD |
| Stacey (1963) | 1 | 1 | MD |

A summary of the predictions for both $S_{ai}/S_d$ and $S_{ar}/S_d$ given by various magnetic viscosity theories is shown in Table 1. An implicit assumption in all the VRM theories is the complete symmetry between the relaxation spectra for acquisition and decay. The factor of two difference in Neel's MD theory comes from the expressions for the Rayleigh constants and not from an implicit difference in the acquisition and decay spectra. The Richter theory can, however, give ratios of $S_{ai}/S_d$ different from 1 if the relaxation spectra for acquisition and decay are different (see fig. 1 in Dunlop 1973). Walton (1980) generalizes the Richter theory by assuming a power series distribution of particle volumes and suggests that $S_a$ may have little relationship to the corresponding $S_a$, although no explicit expressions for $S_{ai}/S_d$ are given.

### 2.2 DIFFUSION AFTER-EFFECT AND DISACCOMMODATION

Diffusion after-effect and disaccommodation have also been the subject of numerous studies for the past 50 years (see review by Rathenau & de Vries 1969). Relaxation equations for the time change in susceptibility (disaccommodation), or magnetization (diffusion after-effect), are of the general form:

$$\chi(t) = \chi_{eq} - (\chi_{eq} - \chi_0) \exp(-t/\tau_1) \tag{7a}$$

$$J(t) = J_{eq} - (J_{eq} - J_0) \exp(-t/\tau_2) \tag{7b}$$

where $\chi_0$ and $J_0$ are initial values at $t = 0$ and $\chi_{eq}$ and $J_{eq}$ are the equilibrium values for $t \gg \tau_1$ or $\tau_2$. $\tau_1$ and $\tau_2$ are characteristic relaxation times for the diffusion process (Néel 1951; Brissonneau 1958; Trukhin 1972) and $\tau = A \exp(E_d/kT)$. $E_d$ is the activation energy for the diffusing species and $A \approx 10^{-13}$ s for magnetite (Kronmüller *et al.* 1974).

Typical values for $E_d$ for vacancy and cation diffusion in ferrites average around 1.0 eV (e.g. Krupicka & Zaveta 1968; Kronmiiller *et al.* 1974). Relaxation times in magnetite ranging from $10^2$s to $10^4$yr are calculated for $0.8 < E_d < 1.2$ eV. Diffusional processes, therefore, have mostly short relaxation times and may not contribute greatly to long-term ($> 10^6$yr) viscous behaviour. Relaxation times, however, are within experimental times and could contribute to viscous behaviour observed in the laboratory.

Equations (7a—b) can be generalized for a spectrum of relaxation times and a logarithmic. time dependence may be derived (Richter 1937). In this respect, the time dependence for susceptibility or magnetization due to diffusion is similar to the time dependence due to thermal fluctuations (Street & Woolley 1950). The important difference is that the activation energies for diffusion are determined by the diffusing species (cations, vacancies, electrons) and, in all probability, are quite different from those produced by thermal fluctuations. Both effects probably occur simultaneously over certain time and temperature intervals, however.

Diffusion after-effect and disaccommodation also exhibit a pronounced temperature dependence (Street & Woolley 1949; Krupicka & Zaveta 1968; Kronmuller *et al.* 1974).

Studies on magnetite and other ferrites suggest that at very low temperatures, electron hopping between $Fe^{+3}$ and $Fe^{+2}$ ions on octahedral lattice sites and electron tunnelling are responsible for the observed disaccomrnodation (Egami 1973; Kronmiiller & Waltz 1980). Room temperature disaccomrnodation is attributed to vacancy diffusion; high temperature disaccomrnodation is related to cation diffusion (Yanase 1962; Krupicka & Zaveta 1968; Kronmiiller *et al.* 1974). The temperature dependence of disaccomrnodation can, therefore, be very complex.

Susceptibility disaccomrnodation spectra are usually determined by measuring $x$ isothermally at two different times after demagnetization and plotting the results as a function of temperature. Disaccomrnodation spectra for magnetite and titanomagnetite are shown in Fig. 1. Disaccomrnodation is clearly characterized by well-defined maxima, suggesting narrow ranges of relaxation times. The possibility exists, however, that over extended periods the relaxation times related to the high temperature peaks could contribute to disaccomrnodation at lower temperatures. The temperature dependence of disaccomrnodation shown in Fig. 1 is much different from the temperature dependence of the viscosity coefficient. In the latter case, $S_a$(or $S_d$) usually increases monotonically with temperature (e.g. Shimizu 1960; Dunlop 1983) indicating a broader and more continuous distribution of relaxation times.

Disaccomrnodation at room temperature and above is dominated by vacancy diffusion. In particular, as the number of vacancies increase, the amount of disaccomrnodation also increases (Yanase 1962; Kronmiiller era/. 1974;Skovorodkin,Tikhonov & Bezuglaya 1975). Kronmiiller *et al.* (1974) suggest that it is only octahedral (B-site) vacancies and ferrous ions that contribute to disaccomrnodation in magnetite. Disaccommodation in a slightly titaniferrous magnetite ($x$ = 0.025) was studied by Hohne *et al.* (1975). The disaccomrnodation in the titanomagnetite was markedly reduced with respect to magnetite (Fig. 1). It is important to note that most ferrites studied are only slightly non-stoichiometric ($z < 0.1$, where $z$ is the oxidation parameter of O'Reilly & Banerjee (1967)); it is unknown if the effects are enhanced, or even persist in the more vacancy-rich titanomaghemites found in nature.

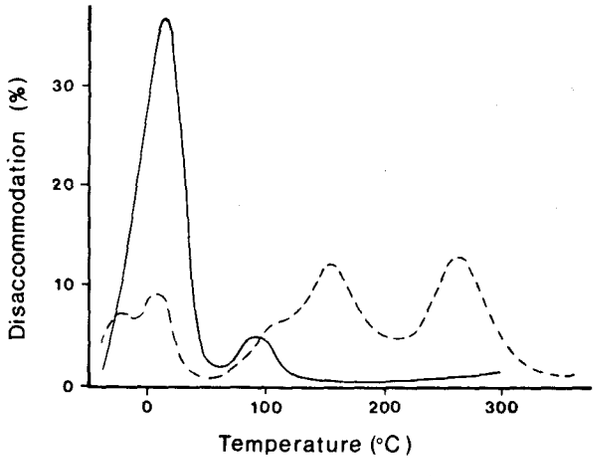


**Figure 1.** Disaccommodation spectra for magnetite (solid line) and titanomagnetite ($x$ = 0.025) (dashed line). Data points are obtained by measuring susceptibility ($\chi$) at $t$ = 30 s and again at $t$ = 300 s following demagnetization by an alternating magnetic field. The parameter $[\chi(30) - \chi(300)]/\chi(30)$ is then plotted as a function of temperature. The figure is adapted from Höhne *et al.* (1975).

In ferrites, such as magnetite and titanomagnetite, disaccommodation at or slightly above room temperature is attributed to the diffusive reordering of octahedral vacancies and ferrous ions. Only in ideal substances devoid of point defects, vacancies, dislocation, etc., would disaccommodation and diffusion after-effect be negligible. The degree of non-stoichiometery is therefore an important parameter to know, in addition to grain size, initial state, *dt*, etc., when comparing viscous behaviour between different samples. These departures from stoichiometry can be very slight ($z < 0.1$) to produce significant amounts of disaccommodation (e.g. Kronmiiller *et al.* 1974). For synthetic and natural magnetites, these small degrees of non-stoichiometry are extremely difficult to measure; nevertheless, they probably contribute to the conflicting evidence about the effects on VRM produced by different initial states of magnetization. It may be possible, however, to use disaccommodation experiments to estimate the degree of non-stoichiometry in natural magnetites if a suitable set of calibration samples can be obtained.

Disaccommodation and diffusion after-effect are competing processes. Diffusion after-effect produces domain wall displacement, while disaccommodation produces domain wall stabilization. Once a wall is displaced, a new local redistribution of vacancies (or cations) occurs producing an increase in the height of the potential barriers that the wall will eventually have to overcome. This will gradually cause the wall to slow down until complete stabilization occurs. This, however, does not preclude thermal activation of the wall leading to a new round of vacancy redistribution. Therefore, when disaccommodation is observed, one cannot easily separate diffusional and thermal fluctuation after-effects by simply measuring $S_a$ or $S_d$.

## 3 Samples

VRM was measured on some oceanic basalts from IPOD site 417D. This site is located approximately 640 km south of Bermuda, and situated in the Mesozoic magnetic anomaly MO (~ 108 Ma). The recovered basalts include fine-grained pillow basalts and coarse-grained massive flows. The magnetic properties of these two groups of samples are distinctly different and are discussed elsewhere (Moskowitz & Banerjee 1981). The samples are oxidized with Curie temperatures between 250 and 450°C; hysteresis and remanence measurements indicate that the magnetic carriers in these basalts are pseudo-single domained (Moskowitz & Banerjee 1981).

## 4 Experimental procedures

Remanence was measured using a spinner magnetometer. Situated within the mu-metal shields of the spinner was a small coil capable of producing DC fields < 10 Oe. A typical experiment usually consisted of the following: (1) the sample was AF demagnetized at 1000 Oe; (2) a 1.0 Oe field was applied for an interval of time dependent on the total duration of the experiment (5 min intervals for the 1 hr experiments, longer intervals for the longer-term experiments); (3) the field was removed and the remanence was measured (*t*~ 15s); (4) steps (2) and (3) were repeated until the acquisition experiment was finished. The decay of this remanence was measured at regular intervals immediately following the acquisition experiment. The acquisition experiments measured a VRM, which differs from the VIM measured by Tivey & Johnson (1981, 1984), Prevot (1981) and Dunlop (1983). The data for the 1 hr acquisition and decay experiments were taken from Moskowitz & Banerjee (1981).

The experimental VRM data were fitted by least squares to linear and quadratic log $t$ functions

$$J_{vrm} = a_o + S \log t \tag{8a}$$

$$J_{vrm} = a_1 + S \log t + b(\log t)^2 \tag{8b}$$

where $a_o$, $S$, $a_1$, $a$ and $b$ are constants. If the least squares fit to (8b) was not significant at the 5 per cent level, then (8a) was retained.

The initial state of magnetization prior to the VRM experiments was varied by storing samples in a residual field $< 100\,\gamma$ for various lengths of time ($\Delta t$) after AF demagnetization at 1000 Oe. A measure of the disaccommodation or stabilization of magnetization was determined by comparing the remanence of a 2 hr VRM with and without zero field storage (Moskowitz & Banerjee 1981):

$$\text{DA} = [J_{vrm}(t, 0) - J_{vrm}(t, \Delta t)]/J_{vrm}(t, 0). \tag{9}$$

The $\Delta t$'s range from 2 hr to 1 week. $S_a$ and $S_d$ were also measured in some instances as a function of $\Delta t$.

## 5 Experimental results

### 5.1 ACQUISITION AND DECAY EXPERIMENTS

Typical acquisition and decay curves for some of the oceanic basalts are shown in Fig. 2. Deviations from linear log $t$ behaviour are slight, but significant during the 1 hr experiments. A viscosity coefficient determined from equation (8a), therefore, adequately characterized their average viscous behaviour. Values for $S_{ar}$ and $S_d$ are compared for both the oceanic basalts (Fig. 3a) and some synthetic samples (Fig. 3b). In all cases for the natural samples, the acquisition of VRM was more rapid than its decay ($S_{ar}/S_d > 1$). Dunlop & Hale (1977) observed similar behaviour for basalt samples from Leg 37. On the other hand, the synthetic

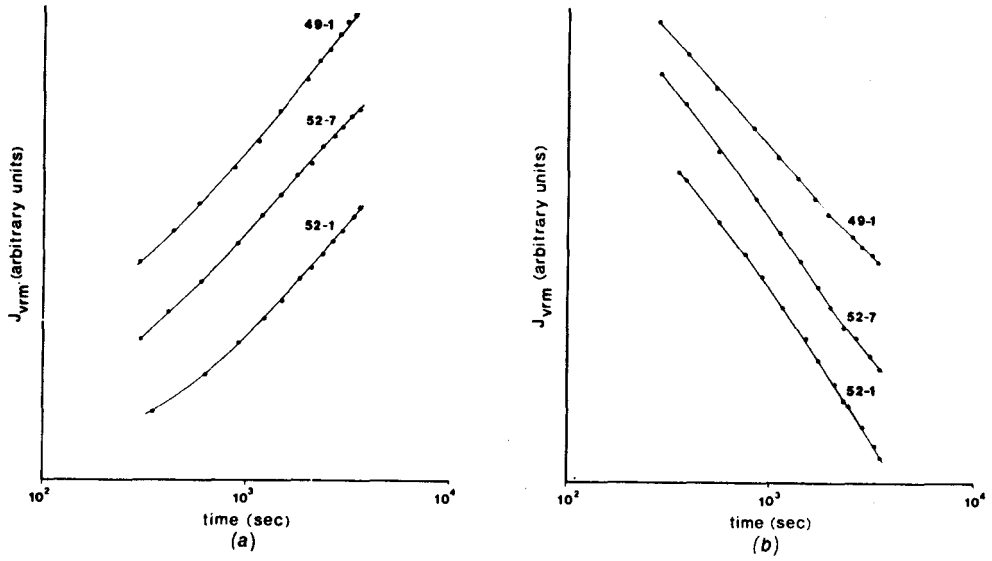


**Figure 2.** Typical (a) acquisition and (b) decay curves for a weak-field (1.0 Oe) viscous remanent magnetization for some oceanic basalts from site 417D. Coarse-grained samples are 49–1 and 52–1 and a fine-grained sample is 52–7.

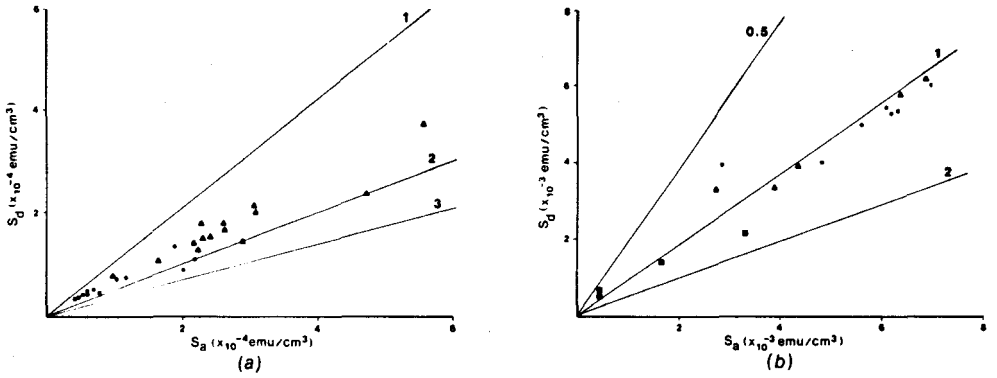


**Figure 3.** Comparison of the corresponding acquisition ($S_{ar}$) and decay ($S_d$) coefficients for (a) oceanic basalts and (b) synthetic samples. In (a) the coarse-grained and fine-grained samples are denoted by triangles and solid dots, respectively, and are taken from Moskowitz & Banerjee (1981). In (b) triangles and solid dots correspond to series 16 and 9 synthetic samples of Moskowitz & Banerjee (1981); squares correspond to synthetic samples of Ozdemir & Banerjee (1981). The numbers on the solid lines correspond to various slopes: $S_a = S_d$, $S_a = 2S_d$, etc.

samples showed an apparent compositional trend. Acquisition of VRM was less rapid than its decay ($S_{ar}/S_d < 1$) for the slightly oxidized samples ($z < 0.1$, see also Table 3) studied by Moskowitz & Banerjee (1981), whereas the more oxidized samples had $S_{ar}/S_d > 1$. The ultrafine grained samples (d~ 0.04/um) studied by Ozdemir & Banerjee (1981) also produced the same trend, but for $z < 0.6$. This effect appears not to be dependent on the ability of the samples to acquire VRM, nor on their grain size. Recent VRM experiments on an unoxidized synthetic titanomagnetite ($x = 0.6$) reported by Sporer (1984), however, gives $S_{ar}/S_d > 1$. The oceanic basalts in the present investigation were all oxidized ($T_c > 250$ C). The only case reported in the literature where both $S_{ar}$ and $S_d$ were measured on a slightly oxidized oceanic basalt ($T_c$~ 180°C, Type I sample of Dunlop & Hale 1977) also had $S_{ar}/S_d > 1$. It is unclear whether the oxidation dependence of $S_{ar}/S_d$ observed for the synthetic samples is fundamental, or perhaps related to sample preparation techniques (e.g. multi-phase samples).

There were no significant correlations between $S_{Bt}/S$^ and grain size, or coercivity, or domain state (as indicated by $J_r/J_s$ and $h_r/h_c$) for either the natural or synthetic samples. This is also observed in magnetite (Dunlop 1983).

### 5.2 NON-LOG *t* BEHAVIOUR

For the natural samples, the 1 hr experiments produced only slight, but significant departures from a linear log *t* curve; however, longer experiments produced markedly quadratic behaviour (Fig. 4). The linear and quadratic terms were positively correlated for both acquisition and decay (Fig. 5), the greater the departure from linear behaviour ($|b|$ large) the larger the value for $|a|$. Also, the ratios of the quadratic terms for acquisition ($b_a$) and decay ($b_d$) were greater than corresponding ratios of $S_{ar}/S_d$. This means that departures from a linear log *t*- behaviour were more pronounced during acquisition than during decay. Similar behaviour is observed for some leg 37 basalts (Plessard & Prevot 1977) and for magnetite (Dunlop 1983).

The fine-grained (~ 1 mkm) synthetic titanomaghemites studied by Moskowitz & Banerjee (1981), in contrast, produced linear log *t* behaviour during aquisition, but quadratic behaviour during decay. However, this may be due to the short duration of the experiments. The

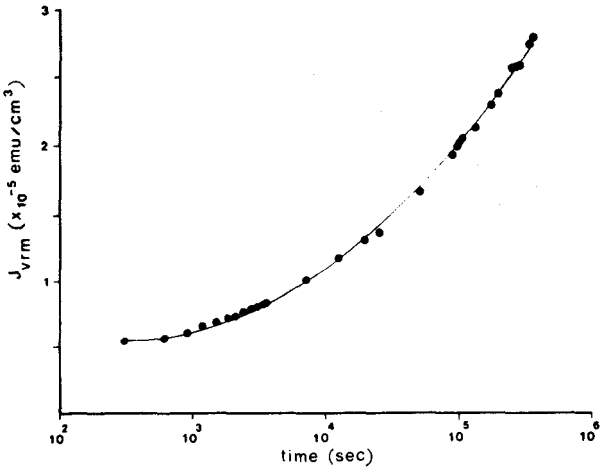


Figure 4. The time dependence of VRM for a coarse-grained sample (50-2) showing markedly non-log time behaviour,

more viscous samples studied by both Moskowitz & Banerjee (1981) and Ozdemir & Banerjee (1981), did produce results similar to those described for the natural samples. The non-log $t$ behaviour of $/_{wm}$, therefore, appears to be a fundamental aspect of VRM.

Experimental VRM observations by Barbier (1951) for a number of oxides and alloys and by Pfrenger & Stierstadt (1967) for nickel indicate that $S_v$ is proportional to $h_c$. Neel's MD model (equation 6b) together with equation (8b) predicts that

$$b/a = S_v/2h \propto h_c/h. \tag{10}$$

This relationship, however, is not substantiated by the present experimental data (for $h$ constant, but variable $h_c$).

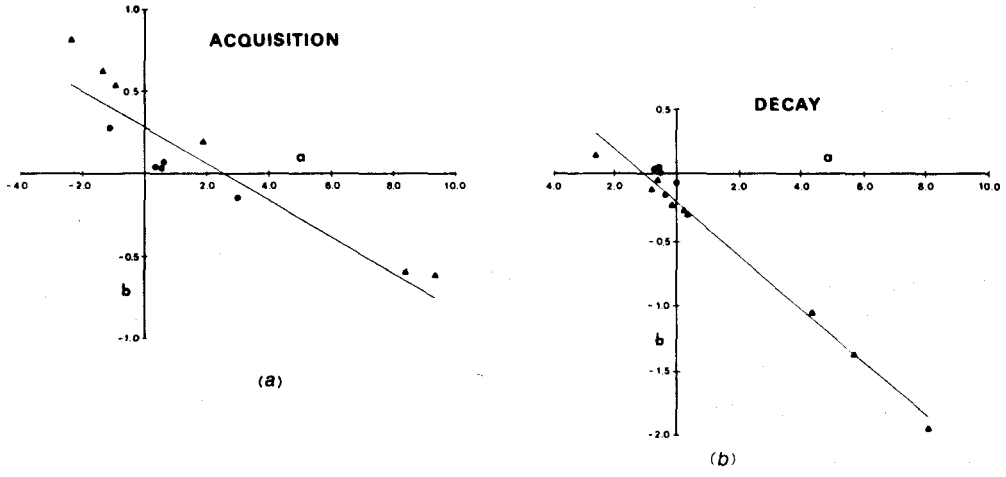


**Figure 5.** The correlation between the linear and quadratic terms in $J_{VRM} = a_1 + a \log t + b (\log t)^2$ for (a) acquisition and (b) decay of VRM. The triangles and solid dots correspond to the coarse-grained and fine-grained oceanic basalts respectively. The solid lines correspond to a linear-least squares fit to the data. Units are in $10^{-5}$ emu cm$^{-3}$.

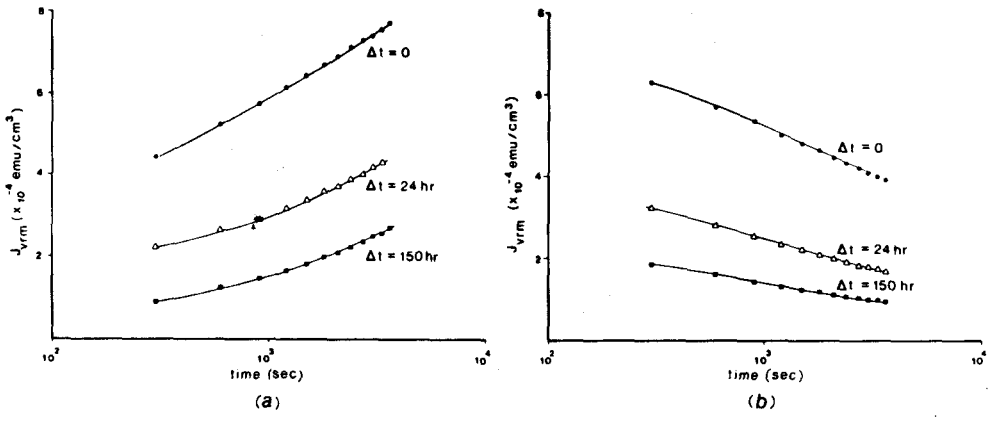


**Figure 6.** Acquisition (a) and decay (b) curves as a function of zero-field storage time ($\Delta t$) following demagnetization. The sample is a coarse-grained massive flow (50–2).

### 5.3 DISACCOMMODATION EXPERIMENTS

The effect of zero field storage time (*dt*) on acquisition and decay of VRM is shown in Fig. 6 for some of the oceanic basalts. $S_{ar}$ and $S_d$ both decreased as Af increased. Similar observations have been made for magnetite (Belokon' & Afremov 1972; Tivey & Johnson 1981, 1984) and synthetic titanomagnetite (Sporer 1984). However, during longer experiments (Fig. 7), *SM* increased with time at a slightly faster rate from the disaccommodated state ($\Delta t = 150$ hr) than from the zero state ($\Delta t = 0$) for $t > 10^4$ s. Given sufficient time ($t_a \gg \Delta t$) the time dependence of VRM should be independent of $\Delta t$, or, in other words, curves of different $\Delta t$ will merge (Ivanov & Belokon' 1976).

The value of $\Delta t$ also had a significant influence on the time dependence of $J_{vrm}$ (Table 2). As $\Delta t$ increased the decay curves became more linear in log $t$ than the corresponding acquisition curves ($b_a/b_d > S_{ar}/S_d$) and overall the linearity in log $t$ for both the acquisition ($(a/b)_a$) and decay ($(a/b)_d$) curves became more pronounced. Similar trends were observed in the acquisition and decay curves presented by Plessard (1971) and Tivey & Johnson (1984).

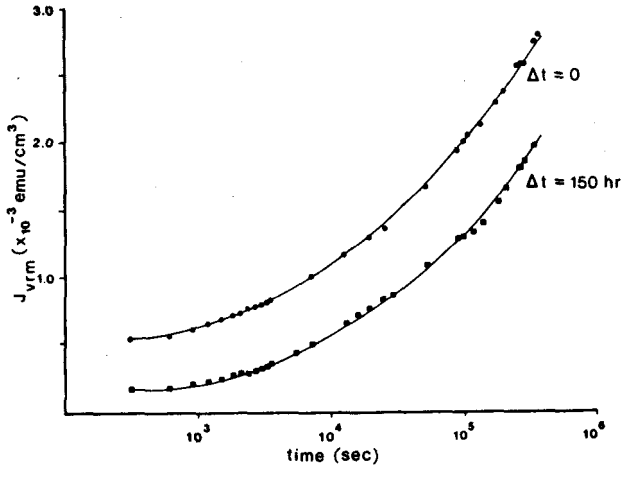


**Figure 7.** VRM acquisition curves for $\Delta t = 0$ and $\Delta t = 150$ hr for a coarse-grained sample (50–2).

**Table 2.** The effect of zero field storage on the time dependence of VRM for a coarse-grained basalt (50–2).

| $\Delta t$ | $S_{ar}/S_d$ | $b_a/b_d$ | $(a/b)_a$ | $(a/b)_d$ |
|---|---|---|---|---|
| 0 | 1.36 | 1.72 | 1.11 | 3.00 |
| 24 hr | 1.39 | 3.48 | 3.82 | 5.64 |
| 150 hr | 1.88 | 9.83 | 4.09 | 16.5 |

The parameter DA defined by equation (9) was used as a measure of disaccommodation. DA increased monotonically with $dt$ (Fig. 8). A very significant amount of disaccommodation, amounting to a change of 40-70 per cent, occurred within just one week of zero field storage after demagnetization.

There were no significant correlations of DA with a number of magnetic parameters. In particular, DA was not correlated with coercivity, susceptibility, domain state, $S_{ar}$ or $S_d$), or with $S_{ar}/S_d$. Nor was it correlated with the parameters describing the time dependence of

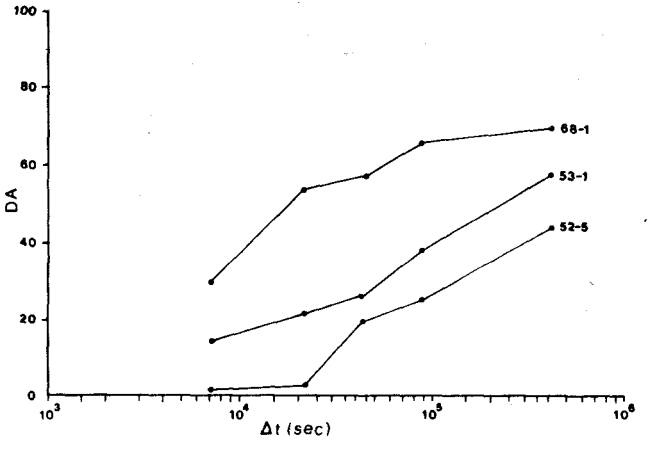


**Figure 8.** DA as a function of $\Delta t$ for coarse-grained (68–1) and fine-grained (53–1, 52–5) samples.

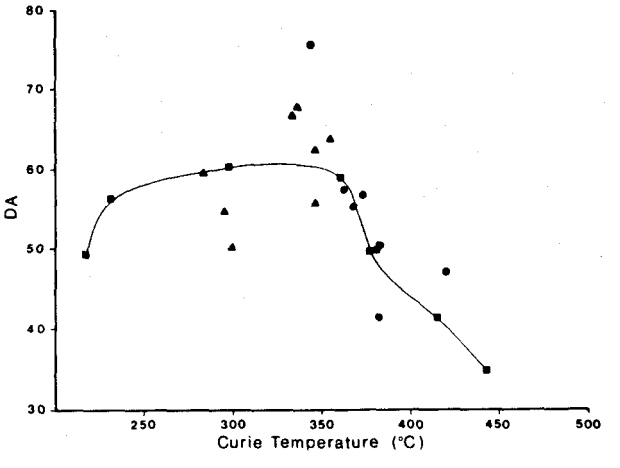


**Figure 9.** DA as a function of Curie temperature. Triangles and solid dots denote the coarse-grained and fine-grained samples, respectively. The smooth curve is for the synthetic titanomaghemites (series 16 of Moskowitz & Banerjee 1981 and denoted by squares).

VRM *{alb).* However, DA did vary with Curie temperature (or degree of oxidation) in a complex manner (Fig. 9). DA decreased with $T_c$ for the natural samples, while DA initially increased, then decreased with $T_c$ for the synthetic samples. Again, because of a lack of unoxidized natural samples, it is uncertain whether the trend for the synthetic samples will be followed by the natural samples.

## 6 Discussion

The experimental VRM data described in the previous section cannot entirely be explained by thermal fluctuation after-effect alone. In this section I will discuss these results and, where appropriate, existing experimental data on magnetic after-effect and disaccommodation to show how they might complete the explanation. This section will concentrate primarily on: (1) comparison of acquisition and decay coefficients; (2) the inadequacies of Neel's viscosity field theory for MD grains; and (3) the importance and limitations of diffusion after-effects and disaccommodation in explaining certain aspects of VRM.

A summary of experimental data pertaining to the ratio of $S_a/S_d$ obtained from an AF demagnetized initial state for both VRM and VIM is given in Table 3. There are, unfortunately, very few studies where both acquisition and decay data for the same sample are reported. Two important observations from Table 3 are: (1) $S_{ar}/S_d$ does not equal one in many studies, and (2) some SD particles do not have $S_{ai}/S_d = 1$. Some of the synthetic magnetites studied by Tivey & Johnson (1981, 1984) and by Dunlop (1983) have average grain sizes well within the SD range and according to theory should have $S_{ai} = S_d$. Possible explanations for this discrepancy are: (1) magnetostatic interactions (although Tivey & Johnson 1981 see no concentration dependence on VRM properties); (2) grain-size distribution for these samples is not as advertised and there is contamination by larger grain sizes. Both suggestions, however, seem *ad hoc.* The VRM results for lunar samples are particularly interesting on this point. For the few lunar samples studied for VRM it is observed that $S_{ar} = S_d$. The iron particles contributing to the VRM in these lunar samples are believed to be nearly superparamagnetic, equi-dimensional, and non-interacting (e.g. Nagata & Carleton 1970; Dunlop *et al.* 1973) and may actually fulfill the assumptions of the Richter and Neel model for SD particles.

Yakubaylik (1968) measured both $S_{ai}$ and $S_d$ for MD magnetite as a function of the demagnetizing factor, A', in order to investigate the influence of the internal demagnetizing field on VRM. He observed that as W increased: (1) $S_{ai}$ decreased; (2) $S_d$ increased; (3) $S_{ai}/S_d$ decreased; and (4) $S_{ai}/S_d < 1$. The variation of $S_{ai}$ with $N$ agrees with the theoretical predictions of Street, Woolley & Smith (1952) and Stacey (1963). The $S_d$ variation, however, is unaccounted for by any VRM theory. Dunlop (1973) suggests that the influence of the internal demagnetizing field should accelerate viscous decay, but retard acquisition. Yakubaylik's data corroborates this notion, but other data do not (Table 3). Furthermore, as $N$ approaches zero $S_a$ becomes greater than $S_d$ (Yakubaylik 1968). In other words, by neglecting the internal demagnetizing field, the decay of VRM becomes slower than its growth. This certainly appears to be the experimentally observed trend (Table 3). It is clear, however, that the experimental results for $S_{ar}/S_d$ and $S_{ai}/S_d$ obtained on a wide variety of SD and MD particles do not agree with the predictions of any VRM theory.

VRM behaviour in MD particles is not well explained by Neel's MD model. Prevot (1981) and Dunlop (1983) demonstrate that Neel's theoretical predictions for AF and thermal demagnetization of VRM are inconsistent with their experimental evidence. In general, non-log $t$ behaviour of $J_{wm}$ can be reconciled with Neel's MD formulation (e.g. equation 6), however, the experimental results of this investigation do not show the specific relationship

**Table 3. Experimental values for $S_{ar}/S$^ and $S_a$^/S^.**

| Material | $S_{gr}/S_d$ | $S_{ai}/S_d$ | ref. |
|---|---|---|---|
| oceanic basalts | | | |
| leg 52 | | | |
| pillow basalts | 1-2.5 | | Moskowitz & Banerjee (1981) |
| massive flows | 1-2.5 | | Moskowitz & Banerjee (1981) |
| leg 51 | | | |
| doleritic basalt | | 1.26 | Smith (1984) |
| leg 37 | | | |
| Type I, II | >1 | | Dunlop & Hale (1977) |
| pillow basalts | | 1.2-1.8 | Plessard & Prlvot (1977) |
| Subaerial basalts | | 1-2 | Prevot (1981) |
| Syn. Titanoraagnetites | | | |
| <d> ^ .04 pm, z<0.6 | <1 | | Ozdemir & Banerjee (1981) |
| <d> i 04 pm, z>0.6 | >1 | | Ozdemir & Banerjee (1981) |
| <d> i, 1 pm, z<0.1 | 0.7-0.85 | | Moskowitz & Banerjee (1981) |
| <d> -\. 1 $_M$m, z>0.1 | 1.1-1.5 | | Moskowitz & Banerjee (1981) |
| <d> ^ 40 pm, z-0 | 0.75 | | Moskowitz i Banerjee (1981) |
| <d> t 20-200 ym, z-0 | >1 | | Sporer (1984) |
| Magnetite | | | |
| 0.037 $_y$m < d < 0.22 pm | | 1-2 | Dunlop (1983) |
| 2.1 ym < d < 14.3 pm | | 1-2 | Dunlop (1983) |
| <d> ^ 100-125 pm | | U | Dunlop (1983) |
| 0.016 $_y$m < d < 0.35 ym | | ^2 | Tivey & Johnson (1981,1984) |
| <d> 8-44 ym | | ^,2 | Tivey & Johnson (1981,1984) |
| <d> < 60 pm | | 1,2 | Tivey & Johnson (1981,1984) |
| magnetite ore | -\,1 | *t, l* | Pechnikov (1967) |
| magnetite ore | | <1 | Yakubaylik (1968) |
| sedimentary rocks containing magnetite | <1 | | Trukhin (1966) |
| Other materials | | | |
| $^{Y}$-Fe2°3 (-$^{01}$-$^{0}$-$^{1}$ ("") | +1 | | Dunlop & West (1969) |
| T-Fe. $_9$,Co -$_5$0. (equant) | >vl | | Dunlop & West (1969) |
| Y-$Fe_20$* (soil) | | -v.1 | Le Borgne (1960) |
| Lunar breccia | •>,! | | Dunlop et al. (1973) |
| Lunar breccia | ->,1 | | Gose & Carnes (1973) |
| Lunar breccia | -vl | | Nagata & Carleton (1970) |

predicted by equation (10). Further, equations (4) and (6) predict linear log *t* behaviour for VIM and quadratic log *t* behaviour for VRM, whereas experimentally, almost the exact opposite is observed (Plessard & Prevot 1977; Dunlop 1983).

Where theoretical predictions of Neel's MD model and experimental data are at odds the following explanations have been proffered: (1) the basic assumption of a uniform distribution of particle properties is incorrect (e.g. Walton 1980; Dunlop 1983); (2) there has been contamination by SD particles (i.e. VRM in MD particles is insignificant) (Belous, Tropin & Stretskul 1972; Bol'shakov 1975; Dunlop 1983); and (3) other causes of after-effects are

contributing to VRM, viz., diffusion after-effect (Tropin & Vlasov 1966; Prevot 1981; Sporer 1984).

Attributing VRM solely to diffusion after-effects also has its weaknesses (Sholpo, Belokon' & Sholpo 1972). It is suspicious that in most experimental studies on the diffusion after-effect the possibility of contributions from thermal fluctuations is never mentioned. Because thermal fluctuations and diffusion after-effects in MD particles are described by the same theoretical formulation, separating the two time effects becomes problematical.

Disaccommodation almost certainly contributes to the effect of the zero field storage time on VRM, or the *dt* effect (Tivey & Johnson 1981, 1984). Disaccommodation has been shown to produce comparable effects to the *dt* effect in metals, alloys and ferrites. Therefore, it is likely that the A? effect observed in both synthetic and natural samples of magnetite (Sholpo 1966, 1967; Tivcy & Johnson 1981, 1984), titanomagnetite and titanomaghemite (Plessard 1971; Moskowitz & Banerjee 1981; Sporer 1984) is also due to disaccommodation.

Diffusion after-effect and disaccommodation are usually associated with the interactions between defects and domain walls. When we consider SD particles, however, the mechanisms for disaccommodation are unclear. Street & Woolley (1950) observed disaccommodation in Alnico (a Fe-Al-Ni-Co alloy), which is supposedly composed of SD particles. The *dt* effect observed by Tivey & Johnson (1981, 1984) occurs in both MD and SD particles of magnetite. The diffusion of defects produces local perturbations in the heights and shapes of potential barriers to changes in magnetization through magnetocrystalline and magnetoelastic interactions with spin moments. It would be reasonable to expect these defect-spin interactions to occur in SD particles producing a rotation of SD moments, as well as in MD particles producing domain wall translation. However, as grain size decreases, the number of defects should decrease and thermal fluctuations should eventually dominate. Intuitively, then, one would expect defect-spin interactions to be more pronounced in MD than in SD particles. Krupicka & Gerber (1960) did indeed observe disaccommodation to decrease markedly in Mn-ferrite between a sintered pellet (unknown grain size) and a powder made from this pellet (~ 1 mkm).

Disaccommodation should also inhibit the growth of VRM and produce a decrease in $S_a$ with time. For most experimental times (minutes to days) the opposite is usually observed, but longer experiments ($t \sim 10^3$hr) on some oceanic basalts do show a slowing down of VRM acquisition (Lowrie & Kent 1978). Likewise, disaccommodation should lead to a decrease in *S&* with increasing $t_a$. This behaviour is observed in two synthetic titanomagnetites (Sporer 1984) and in some sedimentary rocks containing magnetite (Trukhin 1966). In contrast, Barbier (1951) (for a number of oxides and alloys), Creer (1957) (for SD hematite) and Dunlop & Hale (1977) (for an oceanic basalt) do not see any variation in $S_d$ with $t_a$.

Disaccommodation experiments on the oceanic basalts (this study) and some synthetic samples (Moskowitz & Banerjee 1981) did not show any consistent correlations with VRM parameters ($S_{ar}/S_d$, $a/b, b_a/b_d$) or with other magnetic parameters. This suggests that either (1) disaccommodation and VRM are two independent effects, or (2) the DA parameter of equation (9) may be an inappropriate measure to use for comparison with VRM properties. A more meaningful parameter would probably be obtained by measuring the time dependence of susceptibility following demagnetization and calculating a 'viscosity coefficient' from $dx/d \log t$.

The disaccommodation data did correlate with Curie temperature and by implication with degree of oxidation (Fig. 9), although not convincingly. It has been suggested that the mechanism of disaccommodation in magnetites at room temperature or slightly above is associated with the number of octahedral $Fe^{+2}$ ions (i.e. degree of oxidation) and their arrangement on these sites. The vacancy concentration governs the number of rearrangements of octahedral ferrous ions which can be effected, and thus controls the magnitude of

disaccommodation. Tetrahedral vacancies and ferrous ions, on the other hand, do not contribute to disaccommodation at all (Yanase 1962; Krupicka & Zaveta 1968; Kronmiiller *et al.* 1974). The initial increase in DA with oxidation observed in the synthetic titanomaghemites (Fig. 9) is probably due to the increase in the number of octahedral site vacancies, while the decrease in DA with continued oxidation probably reflects the decrease in octahedral ferrous ions available for rearrangement. DA should be zero therefore, when.there are no longer any octahedral ferrous ions present.

In a recent study by Bleil & Petersen (1983) on a number of oceanic basalts they propose a two-stage model of oxidation (their model B) incorporating Fe-migration from the lattice. Stage 1 is characterized by oxidation of octahedral $Fe^{*2}$ that continues until z = 0.6. This stage produces vacancies only on octahedral sites. Oxidation continues in stage 2 by oxidation of tetrahedral $Fe^{+2}$ and producing vacancies on these sites. In addition, during stage 2 the remaining octahedral $Fe^{+2}$ migrates out of the lattice and is transported elsewhere. The transition between stages 1 and 2 occurs approximately at z = 0.6, which corresponds to $T_c$ = 250°C (Bleil & Petersen 1983). Because in stage 2, the amount of octahedral $Fe^{+2}$ decreases while the octahedral vacancy concentration remains constant one would predict that DA should decrease with continued oxidation during stage 2. This is exactly what is experimentally observed (Fig. 9) for the oceanic basalts (most of the samples have $T_C$> 250°C and therefore should be in stage 2).

This evidence, however, should not be interpreted as confirming Bleil & Petersen's (1983) oxidation model B. The synthetic samples, which are assumed to have Fe/Ti = constant during oxidation (no Fe-migration), also show the identical behaviour with respect to DA as the oceanic basalts for $T_C$>250°C. At this juncture, therefore, the DA data do not support either the Fe/Ti = constant or the Fe-migration models for oxidation. In addition, there may be two other causes for the decrease in DA with continued oxidation. (1) There is both theoretical and experimental evidence that suggests that there is a change in domain structure from an MD to an SD state during oxidation in oceanic basalts (e.g. Moskowitz 1980; Dunlop 1981). If this mechanism is operative, DA should decrease just because disaccommodation is primarily an MD process. (2) Multi-phase oxidation products would, certainly contribute to a change in VRM characteristics, although it is unknown whether they would also cause DA to decrease with increasing oxidation.

## 7 Conclusions

The experimental results from this study suggest that certain aspects of VRM cannot be explained entirely by existing models of magnetic viscosity due to thermal fluctuations. These include: (1) the Af effect and the influence of the initial state of magnetization on VRM behaviour, (2) non-log $t$ behaviour of $J_{vrm}$, and (3) the ratio of $S_a/S_d$. This does not imply, however, that those VRM data not consistent with thermal fluctuation models must then be attributed to diffusional after-effects. The present experimental data are insufficient adequately to separate the contributions of these two types of magnetic after-effects. Nevertheless, it does appear that both the $dt$ effect and the influence of the initial state of magnetization on VRM behaviour observed in magnetite and titanomagnetites can be attributed to susceptibility disaccommodation produced by a diffusive reordering of vacancies and ferrous ions on octahedral lattice sites. This is supported by a vast amount of data reported for magnetic oxides, alloys, and metals, and by inference, should also apply to magnetite and titanomagnetite. The non-log $t$ dependence of $J_{vrm}$, and the ratios of $S_a/S_d$ do not lend support to any particular model of MD viscosity (thermal or diffusional).

A primary goal of this paper is to acquaint the reader with theoretical and experimental

results on diffusion after-effect and disaccommodation and to show how these results could provide additional insights into VRM. This approach has produced more questions than answers and more experimental research must be performed before these issues can be resolved. The following is a list of tentative conclusions and possible areas where research should be focused:

(1) MD theories of VRM are inadequate in explaining many aspects of viscous behaviour. The only prediction common to all MD theories that is experimentally confirmed is the generally log $t$ dependence of $J_{vrm}$,. In particular, there is a growing body of experimental evidence that does not support Neel's viscosity field theory for MD particles.

(2) It has been assumed by some authors (e.g. Plessard & Prevot 1977; Prevot 1981; Sporer 1984) that over laboratory time-scales ($10^1$— $10^7$s), diffusional after-effect is a factor contributing to the conflicting results between experimental evidence and theoretical predictions based on thermal fluctuations. There is as yet no real evidence to support this assertion.

A parameter that should be measured is $\partial\chi/\partial \log t$, which is solely due to diffusional processes. This 'viscosity coefficient' should then be compared to other VRM parameters fully to ascertain the importance of diffusional after-effect and disaccommodation on VRM behaviour.

(3) Disaccommodation is probably the underlying mechanism contributing to the *dt* effect and the dependence of VRM on the initial state of magnetization. The susceptibility disaccommodation is related to the diffusive reordering of vacancies and ferrous ions on octahedral sites. The vacancy concentration is, therefore, an important parameter that can influence VRM behaviour.

(4) Over geologic time, thermal fluctuations are probably the dominant cause of VRM because of the shorter relaxation times for diffusion after-effect. However, during the early stages of low-temperature oxidation of titanomagnetites at mid-ocean ridges, diffusional after-effects may contribute to VRM due to the increase in the number of octahedral site vacancies produced by oxidation. Furthermore, the elevated temperatures of the oceanic crust near the ridge should accelerate the diffusion process and enhance its contribution to VRM.

(5) The influence of diffusional processes on VRM in SD particles is unclear. The effects should be much less in SD than in MD particles. If this is true, then measurements of susceptibility disaccommodation may prove useful as a magnetic granulometry technique.

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