Available online at www.sciencedirect.com

ScienceDirect

Russian Geology and Geophysics 56 (2015) 1642–1651

RUSSIAN GEOLOGY AND GEOPHYSICS

www.elsevier.com/locate/rgg

Impulse response of viscous remanent magnetization: laboratory measurements by a pulse induction system

Ya.K. Kamnev [a], N.O. Kozhevnikov [a,b,*], A.Yu. Kazansky [c], S.M. Stefanenko [a]

[a] *A.A. Trofimuk Institute of Petroleum Geology and Geophysics, Siberian Branch of the Russian Academy of Sciences, pr. Akademika Koptyuga 3, Novosibirsk, 630090, Russia*

[b] *Novosibirsk State University, ul. Pirogova 2, Novosibirsk, 630090, Russia*

[c] *M.V. Lomonosov Moscow State University, Leninskie Gory 1, Moscow, Russia*

Received 24 July 2014; accepted 4 March 2015

Abstract

Transient electromagnetic responses measured in the field or in the laboratory may bear effects of viscous remanent magnetization (VRM) associated with magnetic relaxation of ultrafine grains of ferrimagnetic minerals or superparamagnetism. The behavior of VRM can be studied in time or frequency domain, TDEM measurements being advantageous because they are done in the absence of primary field and owing to broad time range providing high accuracy of VRM parameters. Another advantage is that the rate of viscous decay measured as voltage decay does not need to be corrected for stable and/or slowly decaying viscous component of total remanence. Time-dependent transient responses of viscous decay follow the power law $a \cdot t^{-b}$, where a is the initial emf signal (varying in a broad range) and b is the exponent approaching 1. Laboratory tests with a pulse induction coil system reveal a strong linear correlation of the parameter a with frequency-dependent magnetic susceptibility $\Delta\kappa$ used commonly for constraining the relative abundances of superparamagnetic particles. Such systems are thus suitable for quick measurements of the large number of samples for detection of superparamagnetic (*SP*) particles and quantifying their contribution. The difference of b from 1, though being minor, exceeds markedly its error in estimates from measured data. Simulated TDEM responses of a superparamagnetic ground show both parameters (a and b) to depend on particle volume distribution, which is prerequisite for inversion of time-domain transients to magnetic properties of rocks and soils.

Keywords: viscous magnetization; superparamagnetism; impulse response; frequency-dependent magnetic susceptibility; magnetic granulometry; TEM survey; accuracy

Introduction

Magnetic viscosity, viscous remanent magnetization (VRM), or a magnetic after-effect, is a property of ferrimagnetic materials to respond with a lag to the applied field because of magnetic relaxation. The lag may range from fractions of a second to tens of thousand years (Trukhin, 1973). Viscous magnetization appears in most of ferrimagnetic materials, including rocks, where it most often results from magnetic relaxation of single-domain (SD) grains in ferrimagnetic minerals (Bolshakov, 1996). Ultrafine SD grains with <100 s magnetic relaxation times are called superparamegnetic (SP) and their relaxation behavior is called superparamagnetism (Dormann et al., 1997).

Magnetic properties of ultrafine grains in ferromagnetic and superparamagnetic minerals have implications for paleoclimates and for the history of natural and cultural alteration of sediments and soils (Bazhenov et al., 2014; Chang and Kirschvink, 1989; Deng et al., 2005; Heller and Evans, 2003; Liu et al., 2004a,b, 2005; Maher, 1998; Maher and Thompson, 1991; Oldfield et al., 1981; Tarduno, 1994; Thompson and Oldfield, 1986; Zhou et al., 1990; Verosub and Roberts, 1995; Xie et al., 2009).

Superparamagnetic particles are abundant in soils affected by human activity, especially ignition (Linford, 2005; Tabbagh, 1986): baked clays, ancient forges, slag and products of its disintegration (Kozhevnikov and Nikiforov, 1995, 1999; Kozhevnikov et al., 2001, 2003). They are also found in natural high-temperature rocks, such as lavas (Eick and Schlinger, 1990), continental flood basalts or traps (Kazansky et al., 2012; Kozhevnikov and Snopkov, 1995; Stognii et al., 2010), and tuffs (Zakharkin et al., 1988), and in magnetite

* Corresponding author.
E-mail address: KozhevnikovNO@ipgg.sbras.ru (N.O. Kozhevnikov)

1068-7971/$ - see front matter
http://dx.doi.org/10.1016/j.rgg.2015.10.010

ores (Kozhevnikov and Snopkov, 1990). In some cases, horizons with SP grains are used as markers for magnetic stratigraphy of layered volcanic rocks (Eick and Schlinger, 1990; Kozhevnikov and Snopkov, 1990, 1995).

The grain sizes, compositions, and contents of ferrimagnetic minerals are studied using different parameters and methods. The relatively rapid, simple and cheap techniques are dual- or multiple-frequency measurements of magnetic susceptibility or anhysteretic and isothermal remanent magnetization (Worm, 1999). Low-temperature magnetic measurements and Mössbauer spectroscopy are reliable magnetic granulometry tools (Banerjee et al., 1993; Hunt et al., 1995), but the instruments they require are rare in small rock magnetic laboratories while the time-consuming measurements are not suited for quick analysis of large numbers of samples.

Measurements of frequency-dependent magnetic susceptibility for superparamagnetism studies are often performed on a *Bartington Instruments MS2 Magnetic Susceptibility* system with an MS2D dual-frequency sensor (Dearing, 1994). The *Bartington MS2* bridge measures the magnetic susceptibility at low ($f_1 = 465$ Hz) and high ($f_2 = 4650$ Hz) frequencies, and the difference between the respective values corresponds to frequency-dependent magnetic susceptibility: $\Delta\kappa = \kappa_{lf} - \kappa_{hf}$. Percentage frequency dependent susceptibility ($\kappa_{fd}\%$ or $\chi_{fd}\%$) corresponding to the relative contributions of SP grains to the total susceptibility is (Dearing, 1994)

$$FD = [(\kappa_{lf} - \kappa_{hf})/\kappa_{lf}] \times 100.$$

The *Bartington MS2* kappabridge is easy to run, efficient, and can handle small samples. Results obtained on this system are reported in a great number of publications which are impossible to cite in a small paper. However, frequency-dependent magnetic susceptibility as a VRM proxy is highly sensitive to accuracy and requires precise measurements (Hrouda and Pokorny, 2011), especially in the case of weakly magnetic samples. The causes of errors in dual-frequency susceptibility data were discussed by Kozhevnikov et al. (2014).

There is also a more recent and superior AGICO Instruments *Kappabridge*, a world's most sensitive commercially available system for measuring magnetic susceptibility, which has three operating frequencies: 0.976, 3.904, and 15.616 kHz (Pokorny et al., 2006, 2011). However, dual- or even triple-frequency measurements fail to resolve some features in magnetic susceptibility spectra at 125 Hz–512 kHz (Kodama, 2013).

Transient responses of magnetization

Magnetic viscosity can be studied either in frequency or in time domain. Of special interest in this respect is magnetic granulometry based on decay of isothermal remanent magnetization (IRM) J_r after switching off the inducing field, i.e., its transient response (Enkin et al., 2007; Machac et al., 2007; Wang et al., 2010; Worm, 1999).

Paramagnetic minerals and multidomain ferrimagnetic grains are magnetized synchronously with the applied field. The relatively small contribution of superparamagnetic particles is measured in the presence of a high inducing field, which results in significant errors in frequency-dependent magnetic susceptibility (Hrouda and Pokorny, 2011; Kozhevnikov et al., 2014) but time-domain (TDEM) responses of viscous decay are recorded in zero external field and are free from these errors.

Viscous magnetization decay is commonly sampled at small intervals over a large range of delay times spanning at least two orders of magnitude (Wang et al., 2010). Among other things, this improves the quality of inversion to the sizes of single-domain grains.

The remanent magnetization J_r shows a logarithmic dependence on time and the rate of viscous decay is often represented by the viscosity coefficient $S = \dfrac{dJ_r}{d(\log t)}$ (Trukhin, 1973; Yu an Tauxe, 2006), which can be derived from the slope of the viscous curve roughly proportional to the superparamagnetic contribution (Dunlop, 1973).

This method has some limitations though. In the general case, the total remanence J_r at each time t after the inducing field switch-off comprises two components:

$$J_r = J_1 + J_2, \quad (1)$$

where J_1 is the stable remanent magnetization and J_2 is viscous magnetization which decays with time and depends on the size distribution of SP grains.

As we noted, frequency-dependent magnetic susceptibility measurements for estimating the contribution of SP particles are performed in the presence of the inducing field which is much stronger than that induced by superparamagnetism. The situation with TEM responses is somewhat similar: Inasmuch as the difference between J_1 and J_r is within a few percent (Sagnotti and Winkler, 2012; Wang et al., 2010) while the exact J_1 value is unknown, relative error in J_2 may be rather high, especially at late delay times.

Another limitation is that $J_r(t)$ recording begins as late as at 0.3 to 15 s after switch-off, for technical reasons (Wang et al., 2010; Worm, 1999); we assume the earliest time to be $t_0 =$ 0.1 s.

The magnetization J_0 acquired by a population of single-domain grains in an external magnetic field decays exponentially after this field has been removed:

$$J(t) = J_0 \exp(-t/\tau),$$

with τ being the relaxation time constant found as (Néel, 1949)

$$\tau = \tau_0 \cdot \exp(KV/kT), \quad (2)$$

where K is the anisotropy energy, V is the particle volume, T is the absolute temperature, k is Boltzmann's constant, and τ_0 is most often 10^{-9} s. For particles with uniaxial anisotropy (Dunlop, 1983; Dunlop and Özdemir, 1997; Wang et al., 2010; Worm, 1999; Worm and Jackson, 1999),

$$K = \mu_0 H_k J_s/2,$$

where $\mu_0 = 4\pi \times 10^{-7}$ H/m is the magnetic permeability of free space, H_k is the microscopic coercivity of magnetic grains (anisotropy field), and J_s is the saturation magnetization. Thus,

$$\tau = \tau_0 \exp\left(\frac{\mu_0 V J_s H_k}{2kT}\right). \quad (3)$$

For a population of noninteracting single-domain particles, the field $H_k = 2.09H_c$, where H_c is the macroscopic coercivity (Stoner and Wohlfarth, 1948; Wang et al., 2010).

With the limitations on early-time measurements, the time constant $\tau \geq 0.1$ s is assumed for SP grains that affect the magnetization decay. Then, it can be shown with an assumption of $K = 2.5 \times 10^4$ J/m^3 for magnetite (Hrouda and Pokorny, 2011), using equation (3), that grains of $V_{\min} \approx 3 \times 10^{-24}$ m^3 corresponding to $d_{\min} \approx 9$ nm remain “invisible” at room temperature ($T = 300$ K). Note that particles of this size often make a large portion of natural populations of SP grains.

Rate of magnetization decay (impulse response)

Measuring the change rate (time derivative) of magnetization, or impulse magnetization response, instead of magnetization itself allows avoiding the problems discussed above. A pulse induction device for such measurements (Fig. 1) works as follows. It generates rectangular current pulses which pass through the transmitter coil and magnetize samples by the induced magnetic field. After the transmitter current is switched off, the acquired magnetization decays according to (1). At each time t the magnetic flux Φ through the coil is proportional to the total remanence of the sample:

$$\Phi(t) = cJ_r(t) = c(J_1 + J_2(t)),$$

where c is the time-independent coefficient controlled by the geometry and relative position of coils and samples, as well as by the number of turns in a coil, transmitter current, etc.

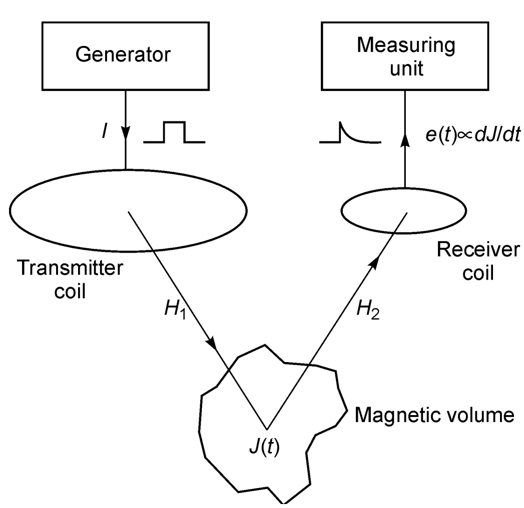

Fig. 1. Pulsed-induction system for measuring VRM transient response.

The time-dependent magnetic flux induces electromotive force (emf) in the receiver coil, which is proportional to the rate of magnetization decay

$$e(t) = -\frac{d\Phi}{dt} = -c\frac{dJ_2}{dt}$$

and does not contain the stable and/or slowly decaying remanence component.

It is convenient to measure the responses of viscous decay with transient electromagnetic (TEM) exploration systems commonly operated in the time range from tens of microseconds to tens or hundreds of milliseconds (Sekachov et al., 2006). Thus magnetic relaxation can be studied at early delay times required for detection of $\leq d_{\min}$ SP particles. Pulse induction systems can resolve SP grains with $\tau \leq 0.1$–1 s against stable single-domain (SSD) particles with very long relaxation times that cause no effect on magnetization decay and multidomain (MD) grains or grains of paramagnetic minerals with short relaxation times that are mute for such systems. Therefore, laboratory measurements of time-dependent magnetization decay are complementary to the rock magnetic methods used for granulometry. Besides the rock magnetic applications, these measurements are useful for interpretation of TEM-TDEM data.

Viscous magnetization is most often treated as geological noise that interferes with TEM responses to be interpreted in terms of “normal” electrical conductivity (Buselli, 1982; Dabas and Skinner, 1993; Lee, 1984a,b; Pasion et al., 2002; Zakharkin and Bubnov, 1995; Zakharkin et al., 1988). The viscous component is most often much weaker than that of eddy current but may be notable or even predominant in some natural (geological) or cultural (archaeological) objects and appear as $1/t$ voltage decay (Fig. 2). Formally interpreted, slow-decaying voltage may be mistaken for a response of a nonexistent conductor (Buselli, 1982; Emerson, 1980), but there are multiple reports that VRM-affected TEM data may bear useful information on the composition, structure and changes of near-surface sediments and soils, as well as on buried manmade objects (Barsukov and Fainberg, 2001, 2002; Kozhevnikov and Nikiforov, 1995, 1999; Kozhevnikov and Snopkov, 1990, 1995; Kozhevnikov et al., 2001; Thiesson et al., 2007).

In this respect, Kozhevnikov and Antonov (2008, 2009, 2011) suggested recommendations of how to increase or decrease the sensitivity of field TEM surveys to magnetic viscosity. Transient responses of a superparamagnetic ground acquired by systems of certain configurations can be inverted in terms of a layered earth with time- or frequency-dependent magnetic susceptibility (Kozhevnikov and Antonov, 2012; Kozhevnikov et al., 2012; Stognii et al., 2010). Thus, TDEM systems are applicable to study 3D magnetic viscosity patterns. Laboratory measurements of viscous decay in samples containing ultrafine ferrimagnetic grains can provide reference for interpretation of field TEM-TDEM data, as well as for design of survey in a potentially superparamagnetic ground.

A laboratory system for measuring VRM impulse response

The experience of measuring VRM impulse response has been limited to the cases of studying the causes of anomalous slowly decaying TEM responses (Buselli, 1982; Emerson, 1980; Kozhevnikov and Snopkov, 1995; Kozhevnikov et al., 2001; Neumann, 2006; Neumann et al., 2005; Zakharkin et al., 1988) or comparing the frequency- and time-domain data (Dabas and Skinner, 1993).

However, the use of such measurements for estimating relative abundances of SP grains or especially their size distributions requires testing laboratory induction systems of different configurations at different delay times on samples of different sizes and magnetizations. As far as we know, no such testing has been undertaken yet.

To bridge the gap, we started systematic laboratory VRM measurements with a pulsed induction coil system configured in different ways. Each following version of configuration and measuring technique based on experience gained in the previous runs. The results were published as short notes as far as they were obtained (Kamnev, 2013; Kamnev et al., 2012, 2013a,b).

The tests were performed using a *FastSnap* TEM device consisting of a transmitter (current switch) and receiver units controlled by a PC via a digital communication line adapter (http://www.sibgeosystems.ru/hardware/FastSnap/). The current switch powered from a 12 V battery generates alternated positive and negative rectangular pulses (max 10 A), at every 20 ms to 1 s, separated by a pause. The voltage induced in the receiver (loop or coil) is sampled at rates of 25 ns to 205 μs, with max 14,000 counts. Preamplified signals from the receiver (loop in field surveys or coil in laboratory tests) are put into a 14-bit ADC. With its dynamic range of 120 dB or more, the receiver can record signal + noise components from fractions of μV to 1 V.

Late-time voltage in such measurements is often underestimated because of short pulse duration (Dabas and Skinner, 1993). Therefore, a correction was applied to bring the measured responses to ideal values corresponding to infinite energizing time.

Step 1. The tests began with choice of samples required to have already known magnetic properties and to show notable time-dependent viscous magnetization. We used core samples of basalt lavas lying over hydrogeneous uranium deposits in Neogene paleovalleys within the Amalat Plateau (Vitim uranium province, Buryat Republic, Russia), where significant magnetic viscosity effects were observed during TEM surveys (Antonov et al., 2011; Kozhevnikov et al., 2012); the magnetic properties of the samples have been documented in detail by Kazansky et al. (2012).

Lava samples, 30 mm high and 63 mm in diameter, were placed into a cylindrical coil of 100 transmitter and 100 receiver turns (Kamnev et al., 2012). The coil size was chosen such that it ensured efficient coupling of the sample with the transmitter and the receiver. Noise reduction was provided by a separate coil making a cancelling system with the receiver.

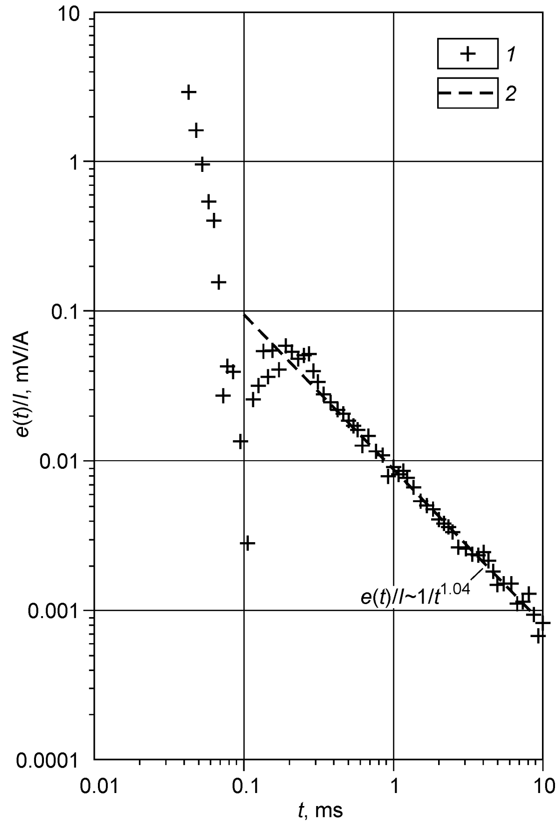

Fig. 2. TDEM response of tuffs measured with a 25 m × 25 m coincident loop by V.A. Vanchugov at Lake Siellyakh in Yakutia. *1*, experimental data; *2*, power-law approximation of late-time response (Stognii et al., 2010).

The rate of viscous decay (Fig. 3*a*) was measured in a time range spanning three orders of magnitude, from $t_1 = 0.1$–0.2 ms depending on the coil's natural response to $t_2 \approx 100$–300 ms (Fig. 2*a*) depending on transmitter current and noise. The range was larger for high-VRM samples.

The current-normalized voltage $e(t)/I$ induced in the receiver shows a power-law decay for all samples:

$$\frac{e\,(t)}{I} = a \cdot t^{-b}, \qquad (4)$$

where a is the signal amplitude (earliest-time voltage) and the exponent b is about 1. This decay pattern, to a high probability, is due to magnetic relaxation of SP grains (Kozhevnikov and Antonov, 2008).

Frequency-dependent magnetic susceptibility was measured at the laboratory of Geodynamic and Paleomagnetism of the Trofimuk Institute of Petroleum Geology and Geophysics with a *Bartington MS2* instrument.

The frequency-dependent magnetic susceptibility (Δκ) and the voltage amplitude (a) measured on lava samples show good correlation (Fig. 3*b*): most points lie near the line $\Delta\kappa = 10 \cdot a$ (dash line in Fig. 3*b*).

Since Δκ is approximately proportional to the volume or weight fraction of SP grains, it is reasonable to infer the same for a, which thus can be used for estimating this amount

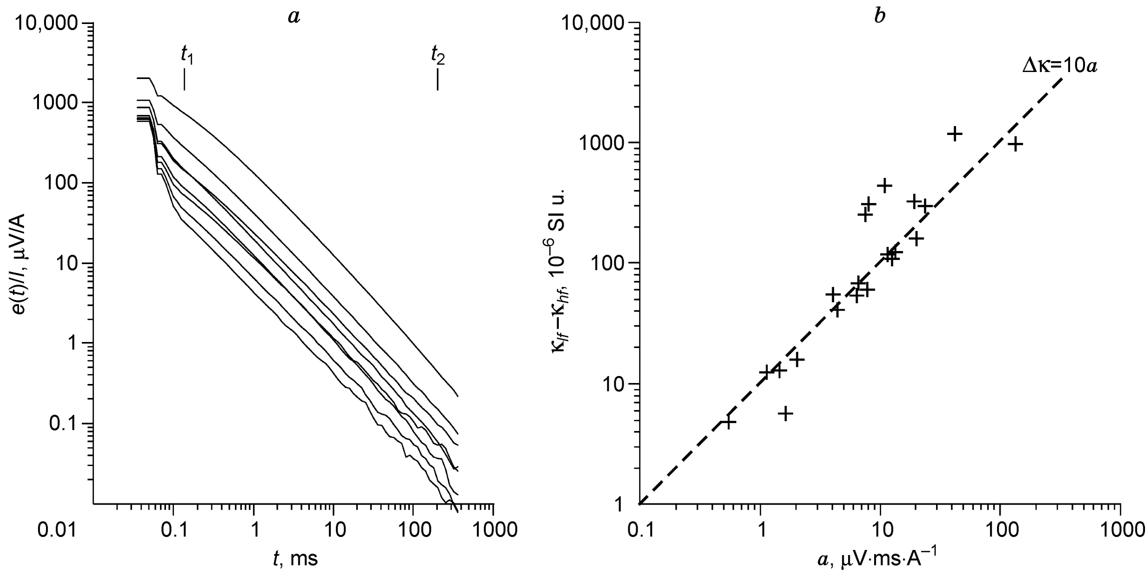

Fig. 3. Impulse VRM responses of basalt samples (*a*); correlation between time- and frequency-domain data (*b*).

varying for about 30 times in the tested samples (Fig. 3*a*). On the other hand, frequency-dependent magnetic susceptibility may be useful to interpret transient responses of a supeparamagnetic ground.

Step 2. The tests of step 1 have demonstrated correlation between time- and frequency-dependent viscous magnetization parameters but highlighted some problems. First, the system of closely spaced multiturn coils has a long (≈ 1 ms) transient self response. This causes no problem as far as they remain much weaker than the signal for samples with high or medium VRM, at late delay times to 200–300 ms, but they obscure the responses of weakly magnetic samples at early times (0.5–1 ms), the signal being as low as noise at >2 ms.

Reducing coil inertia and, correspondingly, minimizing the earliest measurement time, is especially important for small cubic samples (2 cm × 2 cm × 2 cm) commonly used in paleomagnetic studies. Such samples were measured with special cubic coils enveloping the sample tightly to provide the best coupling. However, the transients of most samples were hardly resolvable against noise. We tried to make the coil response shorter by separating the transmitter and receiver coils or to increase the transmitter current and the total recording time (to collect more data for averaging). Both solutions failed as they led to worse coupling in the former case and to heating of samples and uncontrollable drift in the latter case. Note that frequency-dependent magnetic susceptibility is also highly sensitive to minor changes in the sample temperature (Kozhevnikov et al., 2014).

Changing the system configuration was more successful: the receiver coil tightly enveloping the samples was placed inside a larger transmitter coil, and the gap between the two coils was ventilated (Fig. 4) to prevent the samples from heating during longer measurements at stronger current. Furthermore, weaker coupling between the transmitter and receiver coils reduces the duration of the system's transients.

High-frequency (100 kHz–3 MHz) noise, which poses another problem to measurements, was attenuated with a shunt resistor connected parallel to the receiver coil. It acted as a low-frequency bandpass filter with its bandwith proportional to the shunt resistance being 100 Ohm·m at late times and

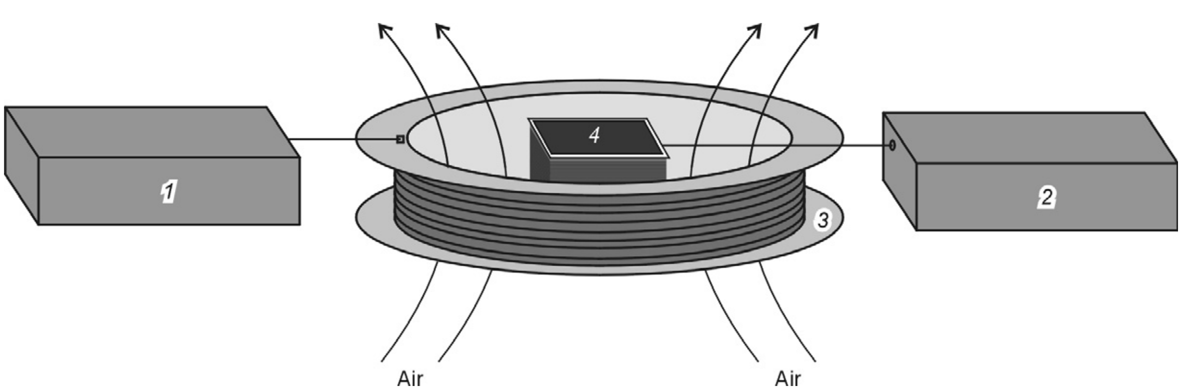

Fig. 4. Sketch of a laboratory pulse-induction system for measuring impulse VRM responses of cubic samples: *1*, generator of rectangular current pulses; *2*, measurement unit; *3*, transmitter coil; *4*, receiver coil.

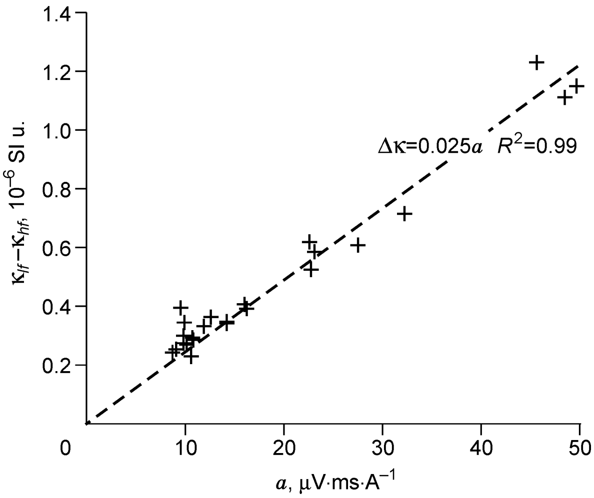

Fig. 5. Correlation between time- and frequency-domain responses of cubic samples cut from walls of an ancient bloomery furnace in Olkhon area.

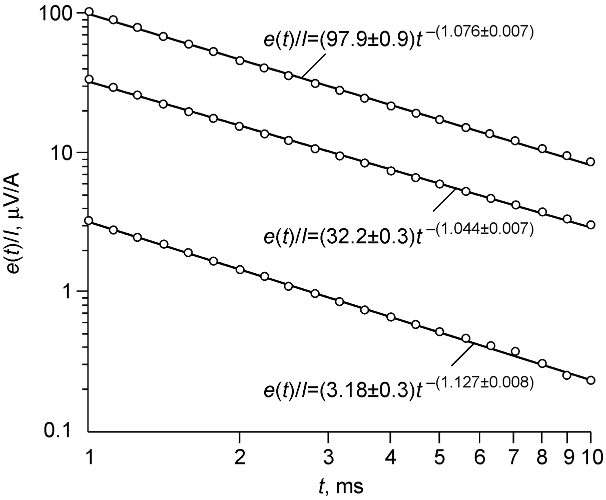

Fig. 6. Impulse VRM responses of magnetically viscous cubic samples cut from walls of an ancient bloomery furnace in Olkhon area.

800 Ohm·m at early times (Kamnev et al., 2013a,b). At 100 Ohm·m shunt, the natural system's response became longer, but it remained stable in laboratory conditions and could be measured at an empty coil without samples and then subtracted from the total response. The high shunt resistance, along with low transmitter current (≈ 0.4 A), reduced the coil response duration to tens of microseconds. Thus the recording time intervals were 60 μs to 70 ms for highly magnetic samples and 1 to 10 ms for samples with low or medium magnetization.

The method was applied to samples with high VRM cut from walls of an ancient bloomery furnace in Olkhon area (Fig. 5) used to smelt iron. Note that the archaeological site of ancient metallurgy was discovered accidentally due to the presence of SP particles in the near-surface soil inferred from TEM data (Kozhevnikov et al., 1998, 2001). The magnetization and composition of ancient slag sampled at the site were studied in detail by Kozhevnikov et al. (2003) but the magnetic properties of the furnace remained unexplored. The transient responses of the furnace wall samples were measured in time and frequency domains (Fig. 5). Dual-frequency measurements of magnetic susceptibility at $f_1 = 976$ Hz and $f_2 = 3904$ Hz were run on an *MFK1 Kappabridge* (Pokorny et al., 2006) at the paleomagnetic center of the Trofimuk Institute of Petroleum Geology and Geophysics. As in the case of the Amalat lava samples (Fig. 3), the frequency-dependent magnetic susceptibility ($\Delta\kappa = \kappa_1 - \kappa_2$) shows good correlation with the voltage amplitude (a). Therefore, pulse induction systems are suitable for measuring viscous decay in standard cubic samples.

Discussion

Measured transient responses of some typical samples with a power-law (4) decay shown in Fig. 6 with errors in the parameters a and b calculated according to (Squires, 1968), differ from one another both in the amplitude (a) and in the exponent (b). The difference in b most likely accounts for the grain size distribution. In this respect, it would seem reasonable to normalize the responses to the initial signal amplitude, but the latter is also related to the grain distribution (see below).

Grains in rocks may be of different sizes and thus have different times of magnetization acquisition/decay, in a range defined by the weight function $f(\tau)$. The distribution of time constants in a population of SD particles with uniformly distributed energy barriers between magnetization states is described by the Frölich function (Fannin and Charles, 1995). The relaxation times τ in this function are in the range from τ_1 to τ_2: $\tau_1 \le \tau \le \tau_2$. Inside the range,

$$f(\tau) = \frac{1}{\tau \ln(\tau_2/\tau_1)} \qquad (5)$$

and outside it $f(\tau) = 0$. With $\ln\tau$ used as argument in (5), the Froelich function becomes $G(\ln\tau) = \frac{1}{\ln(\tau_2/\tau_1)}$, where logarithmic relaxation times are obviously evenly distributed within the range from τ_1 to τ_2.

Exact or "true" τ_1 and τ_2 are commonly unknown but this does not affect the measurement results. The range of time constants normally spans many orders of magnitude while magnetic relaxation is measured at $\tau_1 \ll t \ll \tau_2$. Then, switching on/off a step external field induces magnetization of a population of particles varying as a function of logarithmic time (Kozhevnikov and Antonov, 2008): $J(t) = \frac{\kappa_0 H}{\ln(\tau_2/\tau_1)}(B + \ln t)$, where κ_0 is the static magnetic susceptibility and $B = 1 + \gamma + \ln\tau_2$ ($\gamma \approx 0.577$ is the Euler constant). Correspondingly, $b = 1$ for the voltage decay (4) (Kozhevnikov and Antonov, 2008), which disagrees with the observations (Fig. 6) showing that $b \ne 1$, and the difference depends on the sample.

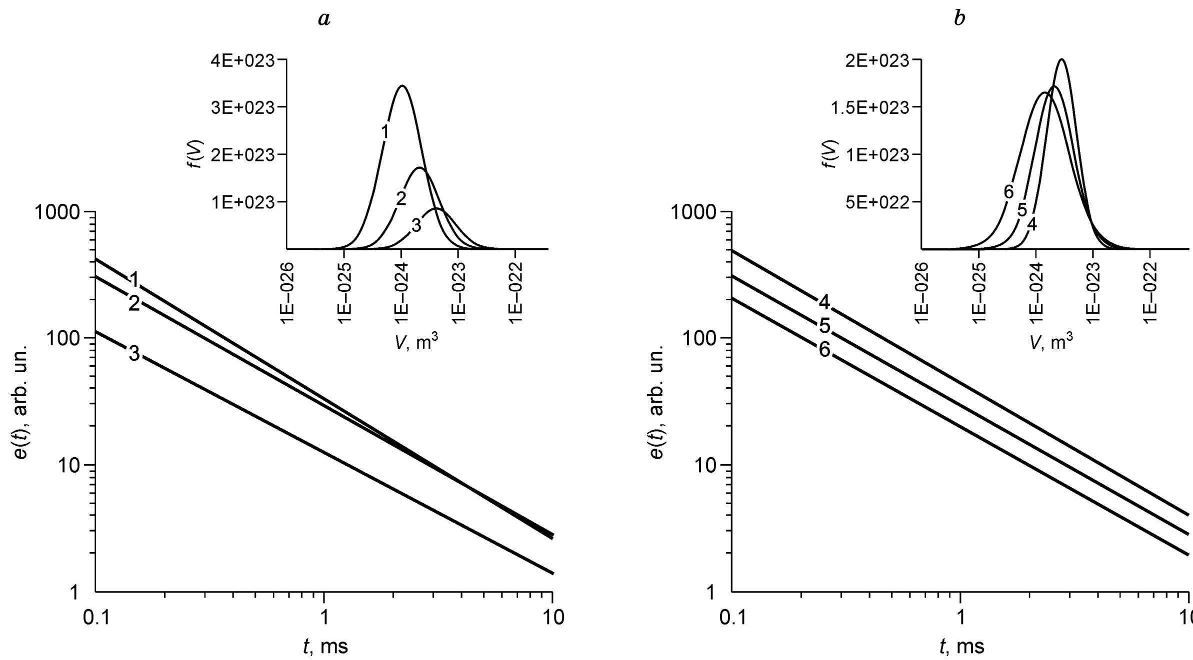

Fig. 7. Impulse VRM responses at different average volumes V_m (*a*) and their standard deviations σ (*b*) (parameters of lognormal volume distribution of single-domain particles).

The exponent *b* may depart from 1 because the relaxation time departs from the distribution given by Eq. (5). This inference is consistent with modeled TDEM responses of a population of SP grains with their relaxation times not fitting the Frölich distribution (Kamnev et al., 2014).

More commonly, the relaxation times of SP grain populations are of secondary importance relative to the distribution of grain volumes and/or diameters. With the particle volume expressed via the time constant τ, according to (2), as

$$V = \frac{kT}{K} \ln(\tau/\tau_0),$$

and knowing how to find the distribution of a parameter depending on other parameter with a known distribution (e.g., Cooper and McGillem, 1998), it is easy to prove that the volumes of particles with the Frölich-law relaxation times are distributed evenly over the interval $V_1 - V_2$:

$$f(V) = \frac{1}{V_2 - V_1},$$

where $V_1 = (kT/K)\ln(\tau_1/\tau_0)$, $V_2 = (kT/K)\ln(\tau_2/\tau_0)$.

We proceeded from the published evidence of lognormal volume distributions of particles found by magnetic granulometry (Banerjee et al., 1993; Eyre, 1997; Kodama, 2013; Liu et al., 2005; Worm, 1999):

$$f(V, V_m, \sigma) = \frac{1}{\sqrt{2\pi}\,\sigma V} \exp\left[-\frac{(\ln V - \ln V_m)^2}{2\sigma^2}\right], \quad (6)$$

where V_m is the average volume and σ is the standard deviation of ln *V*.

To provide more universal results, not restricted to any specific coil system and/or sample, the VRM decay rate $J_2(t)$ of a population of SD grains with lognormal volume distribution was used instead of voltage induced in the receiver. The total VRM is after (Wang et al., 2010)

$$J_r(t) = \frac{\int_0^\infty f(V, V_m, \sigma)\, J_0 \exp(-t/\tau(V))\, dV}{\int_0^\infty V f(V, V_m, \sigma)\, dV}, \quad (7)$$

where J_0 is the initially imposed magnetization. The relaxation time was found by (3) with the parameters for magnetite $\mu_0 H_k = 25$ mT, $J_s = 478$ kA/m (Dunlop and Özdemir, 1997) and $T = 293$ K (room temperature). The initial magnetization J_0 and the average volume of all particles defined by the integral in the denominator of (7) were assumed to be unity, and the emf was thus expressed in arbitrary units.

Modeling shows that both the average volumes V_m and their standard deviation σ influence the behavior of the parameters *a* and *b*, as it is illustrated in Fig. 7. The effect of V_m (Fig. 7*a*) on VRM decay (bottom panel) is shown for three example volume distributions *f*(*V*) given by (6) with the same error (σ = 0.8) but different average volumes $V_m = 2 \times 10^{-24}$, 4×10^{-24}, and 8×10^{-24} m^3 (top panel). The responses for different σ (0.6, 0.8, and 1.0) and the same V_m (4×10^{-24} m^3) are shown in Fig. 7*b*. See Table 1 for the parameters of the distribution and their power law approximation.

Although the volume distribution in Fig. 7 is far from uniform (which is equivalent to the Frölich distribution in terms of τ), the decay pattern almost perfectly ($R^2 = 1$) fits the

Table 1. Parameters of distribution (6) and power-law approximation (4) of voltage decay

Distribution, No.	V_m, 10^{-24} m^3	σ	*a*	*b*	R^2
1	2	0.8	33.2	1.10	1
2, 5	4	0.8	29.5	1.02	1
3	8	0.8	12.5	0.95	1
4	4	0.6	44	1.04	1
6	4	1.0	20	1.01	1

power law (4), while the departure of *b* from unity is small but exceeds the error in the observed values (Fig. 6).

The influence of V_m and σ on the voltage amplitude (*a*) and the exponent (*b*) makes thinking of the possibility of inversion, i.e., finding a distribution of single-domain SP grains fitting the best the observed decay in transient responses. Checking this possibility is beyond the present consideration, but its existence can make basis for a new method of magnetic granulometry of ultrafine grains in ferrimagnetic minerals.

Conclusions

Viscous remanent magnetization (VRM) of rocks affects notably their transient responses measured in the field or in laboratory by a pulse induction system.

Measuring impulse VRM responses is advantageous over frequency-domain studies as magnetic relaxation is measured in the absence of an inducing field while wide time range ensure better accuracy of the VRM parameters.

The rate of decay (impulse response) being free from constant and/or slowly decaying total remanence, there is no need for ambiguous separation of the small VRM component.

The VRM impulse response is described by the power law $a \cdot t^{-b}$, where *a* is the amplitude varying in a broad range and *b* is the exponent approaching 1.

Laboratory pulse-induction measurements of magnetically viscous samples show strong linear correlation of *a* with frequency-dependent magnetic susceptibility Δκ which is commonly used to estimate the contribution of SP grains. Therefore, systems of this kind can provide quick analysis of large numbers of samples to detect SP particles and estimate their relative abundances.

Although being small, the departure of the exponent *b* from unity notably exceeds the standard deviation in measured data. According to forward modeling, the volume distribution of particles influences both parameters (*a* and *b*), which creates prerequisites for inversion, i.e., search for the distribution that fits the best to the observations.

We wish to thank T.I. Perepelova for useful comments.

References

Antonov, E.Yu., Kozhevnikov, N.O., Maksimov, P.S., Gomulsky, V.V., 2011. TEM surveys over magnetically viscous ground, in: Proc. 5th M.N. Berdichevsky and L.L. Vaniyan All-Russian Seminar on Electromagnetic Soundings [in Russian]. EMS-2011. Book 2. St. Petersburg University, St. Petersburg, pp. 11–14.

Banerjee, S.K., Hunt, C.P., Liu, X.M., 1993. Separation of local signals from the regional paleomonsoon record of the Chinese Loess Plateau: A rock–magnetic approach. Geophys. Res. Lett. 20, 843–846, doi:10.1029/93GL00908.

Barsukov, P.O., Fainberg, E.B. 2001. Superparamagnetism effect over gold and nickel deposits. European J. Env. Engineer. Geophys. 6, 61–72.

Barsukov, P.O., Fainberg, E.B., 2002. TEM soundings of environment with regard to IP and SPM effects. Izv. RAN, Ser. Fizika Zemli 11, 82–85.

Bazhenov, M.L., Van der, Voo R., Meert, J.J., Levashova, N.M., Ipat'eva, I.S., 2014. Late Paleozoic geomagnetic-field estimates from paleomagnetic studies of Permian lavas in northeastern Kazakhstan. Russian Geology and Geophysics (Geologiya i Geofizika) 55 (1), 108–117 (133–145).

Bolshakov, V.A., 1996. Rock Magnetism: Applications to Studies of Recent Sediments [in Russian]. Geos, Moscow.

Buselli, G., 1982. The effect of near-surface superparamagnetic material on electromagnetic transients. Geophysics 47 (9), 1315–1324.

Chang, S.B.R., Kirschvink, J.L., 1989. Magnetofossils, the magnetization of sediments, and the evolution of magnetite biomineralization. Annu. Rev. Earth Planet. Sci. 17, 169–195, doi:10.1146/annurev.ea.17.050189.001125.

Cooper, G.R., McGillem, C.D., 1998. Probabilistic Methods of Signal and System Analysis. Series in Electrical and Computer Engineering, Oxford University Press.

Dabas, M., Skinner, J.R., 1993. Time-domain magnetization of soils (VRM), experimental relationship to quadrature susceptibility. Geophysics 58 (3), 326–333.

Dearing, J.A., 1994. Environmental magnetic susceptibility: Using the Bartington MS2 system. Chi. Publ., Fairfield, CT.

Deng, C., Vidic, N.J., Verosub, K.L., Singer, M.J., Liu, Q., Shaw, J., Zhu, R., 2005. Mineral magnetic variation of the Jiaodao Chinese loess/paleosol sequence and its bearing on long-term climatic variability. J. Geophys. Res. 110, B03103, doi:10.1029/2004JB003451.

Dormann, J.L., Fiorani, D., Tronc, E., 1997. Magnetic relaxation in fine-particle systems, in: Prigogine, I., Rice, S.A. (Eds.), Advances in Chemical Physics, Vol. 98, pp. 283–494, J. Wiley & Sons, Hoboken, NJ, doi:10.1002/9780470141571.ch4.

Dunlop, D.J., 1973. Theory of the magnetic viscosity of lunar and terrestrial rocks. Rev. Geophys. 11, 855–901, doi:10.1029/RG011i004p00855.

Dunlop, D.J., 1983. Viscous magnetization of 0.04–100 μm magnetites. Geophys. J. R. Astron. Soc. 74, 667–687.

Dunlop, D.J., Özdemir, Ö., 1997. Rock Magnetism. Fundamentals and Frontiers, Cambridge University Press, New York, London and Cambridge.

Eick, P.M., Schlinger, Ch.M., 1990. The use of magnetic susceptibility and its frequency dependence for delineation of a magnetic stratigraphy in ash-flow tuffs. Geoph. Res. Lett. 17 (6), 783–786, doi:10.1029/gl017i006p00783.

Emerson, D.M. (Ed.), 1980. The Geophysics of the Elura Orebody. Austral. Soc. Expl. Geophys., Sydney.

Enkin, R.J., Baker, J., Nourgaliev, D., Iassonov, P., Hamilton, T.S., 2007. Magnetic hysteresis parameters and Day plot analysis to characterize diagenetic alteration in gas hydrate-bearing sediments. J. Geophys. Res. 112, B06S90, doi:10.1029/2006JB004638.

Eyre, J.K., 1997. Frequency dependence of magnetic susceptibility for populations of single-domain grains. Geophys. J. Int. 129, 209–211.

Fannin, P.C., Charles, S.W., 1995. On the influence of distribution functions on the after-effect function of ferrofluids. J. Phys. D. Appl. Phys. 28, 239–242.

Heller, F., Evans, M.E., 2003. Environmental Magnetism: Principles and Applications of Enviromagnetics. Academic Press, New York.

Hrouda, F., Pokorny, J., 2011. Extremely high demands for measurement accuracy in precise determination of frequency-dependent magnetic susceptibility of rocks and soils. Stud. Geophys. Geod. 55, 667–681.

http://www.sibgeosystems.ru/hardware/FastSnap/

Hunt, C.P., Banerjee, S.K., Han, J., Solheid, P.A., Oches, E., Sun, W., Liu, T., 1995. Rock-magnetic proxies of climate change in the loess-paleosol sequence of the western loess plateau of China. Geophys. J. Int. 123, 232–244.

Kamnev, Ya.K., 2013. Frequency- and time-domain studies of magnetic viscosity, in: Lithospheric Structure and Geodynamics [in Russian]. Proc. XXV All-Russian Youth Conf., Irkutsk, 23–28 April, 2013. Institute of Earth Crust, Irkutsk, pp. 150–151.

Kamnev, Ya.K., Kozhevnikov, N.O., Matasova, G.G., 2012. Laboratory time-domain measurements of magnetic viscosity by a pulsed-induction meter, in: Subsoil Use. Mining. New Lines and Technologies in Mineral Exploration and Development. Proc. Interexpo GEO-Siberia-2012. VIII Int. Congr., 10–20 April 2012, Novosibirsk. SGGA, Novosibirsk, Book 1, pp. 38–43.

Kamnev, Ya.K., Kozhevnikov, N.O., Stefanenko, S.M., 2013a. Laboratory time-domain measurements of magnetic viscosity: method updates, in: Subsoil Use. Mining. New Lines and Technologies in Mineral Exploration and Development. Geoenvironment. Proc. Interexpo GEO-Siberia-2013. IX Int. Congr., 15–26 April 2013, Novosibirsk. SGGA, Novosibirsk, Book 2, pp. 197–202.

Kamnev, Ya.K., Kozhevnikov, N.O., Stefanenko, S.M., 2013b. Time-domain measurements of magnetic viscosity: problems and solutions, in: Proc. 6th M.N. Berdichevsky and L.L. Vaniyan All-Russian Seminar on Electromagnetic Soundings [in Russian]. EMS-2013. Online Publication.

Kamnev, Ya.K., Kozhevnikov, N.O., Stefanenko, S.M., 2014. Forward modeling of TEM responses of magnetically viscous geological materials: First results, in: Subsoil Use. Mining. New Lines and Technologies in Mineral Exploration and Development. Geoenvironment. Proc. Interexpo GEO-Siberia-2014. X Intern. Congr., 8–18 April 2014, Novosibirsk. SGGA, Novosibirsk, Book 2, pp. 101–105.

Kazansky, A.Yu., Matasova, G.G., Kozhevnikov, N.O., Antonov, E.Yu., Maksimov, P.S., 2012. Magnetic after-effect: contribution to transient responses of basalts in the Vitim plateau. Geofizicheskii Zh. 34 (4), 128–136.

Kodama, K., 2013. Application of broadband alternating current magnetic susceptibility to the characterization of magnetic nanoparticles in natural materials. J. Geophys. Res. B118, 1–12, doi: 10.1029/2012JB009502.

Kozhevnikov, N.O., Antonov, E.Yu., 2008. The magnetic relaxation effect on TEM responses of a uniform earth. Russian Geology and Geophysics (Geologiya i Geofizika) 49 (3), 197–201 (262–276).

Kozhevnikov, N.O., Antonov, E.Yu., 2009. The magnetic relaxation effect on TEM responses of a two-layer earth. Russian Geology and Geophysics (Geologiya i Geofizika) 50 (10), 895–904 (1157–1170).

Kozhevnikov, N.O., Antonov, E.Yu., 2011. Magnetic relaxation of a horizontal layer: Effect on TEM data. Russian Geology and Geophysics (Geologiya i Geofizika) 52 (4), 398–405 (512–520).

Kozhevnikov, N.O., Antonov, E.Y., 2012. The TEM method in studies of near-surface magnetically viscous rocks. Proc. Near Surface Geoscience 2012—18th European Meeting of Environmental and Engineering Geophysics, Paris, France, 3–5 September, p. 5.

Kozhevnikov, N.O., Nikiforov, S.P., 1995. Magnetic viscosity of fired clays and the possibility of its use for archaeological prospection, in: Proc. Archaeological Sci. Conf. Bournemouth University, pp. 163–169.

Kozhevnikov, N.O., Nikiforov, S.P., 1999. Magnetic viscosity of fired clay and its archaeological applications. Rossiiskii Geofizicheskii Zh. 13–14, 42–46.

Kozhevnikov, N.O., Snopkov, S.V., 1990. Superparamagnetic Effects in EM Surveys [in Russian]. Deposited in VINITI 13.08.90, N 4584–V90, Irkutsk.

Kozhevnikov, N.O., Snopkov, S.V., 1995. Supermagnetism of traps and its relation to TEM anomalies (Yakutian Kimberlite Province). Geologiya i Geofizika (Russian Geology and Geophysics) 36 (5), 91–102 (89–100).

Kozhevnikov, N.O., Kozhevnikov, O.K., Kharinsky, A.V., 1998. How search for a geophysical solution drove at a discovery of an archaeological site. Geofizika 6, 48–60.

Kozhevnikov, N.O., Kharinsky, A.V., Kozhevnikov, O.K., 2001. An accidental geophysical discovery of an Iron Age archaeological site on the western shore of Lake Baikal. J. Appl. Geophys. 47 (2), 107–122.

Kozhevnikov, N.O., Kozhevnikov, O.K., Kharinsky, A.V., Urbat, M., 2003. Chemical composition and magnetism of ancient metallurgical slags from the Chernorud site on the western shore of Lake Baikal, in: Archaeometallurgy in Europe. Proc. Int. Conf. Milan, Italy, Vol. 1, pp. 525–534.

Kozhevnikov, N.O., Antonov, E.Yu., Matasova, G.G., Kamnev, Ya.K., 2012. The TEM prospection method in studying magnetically viscous ground. Geofizicheskii Zh. 34 (4), 137–149.

Kozhevnikov, N.O., Kamnev, Ya.K., Kazansky, A.Yu., 2014. Error analysis of frequency-dependent magnetic susceptibility measurements: magnetic viscosity studies with the *Bartington MS2* system. Russian Geology and Geophysics (Geologiya i Geofizika) 55 (4), 508–514 (650–659).

Lee, T.J., 1984a. The effect of a superparamagnetic layer on the transient electromagnetic response of a ground. Geophys. Prosp. 32, 480–496.

Lee, T.J., 1984b. The transient electromagnetic response of a magnetic or superparamagnetic ground. Geophysics 49 (7), 854–860.

Linford, N., 2005. Archaeological applications of naturally occurring nanomagnets. J. Phys. Conf. Series, 17, pp. 127–144, doi:10.1088/1742-6596/17/1/019

Liu, Q.S., Jackson, M.J., Yu, Y.J., Chen, F.H., Deng, C.L., Zhu, R.X., 2004a. Grain size distribution of pedogenic magnetic particles in Chinese loess/paleosols. Geophys. Res. Lett. 31, L22603, doi:10.1029/2004GL021090.

Liu, Q.S., Banerjee, S.K., Jackson, M.J., Maher, B.A., Pan, Y.X., Zhu, R.X., Deng, C.L., Chen, F.H., 2004b. Grain sizes of susceptibility and anhysteretic remanent magnetization carriers in Chinese loess/paleosol sequences. J. Geophys. Res. 109, B03101, doi:10.1029/2003JB002747.

Liu, Q.S., Torrent, J., Maher, B.A., Yu, Y.J., Deng, C.L., Zhu, R.X., Zhao, X.X., 2005. Quantifying grain size distribution of pedogenic magnetic particles in Chinese loess and its significance for pedogenesis. J. Geophys. Res. 110, B11102, doi:10.1029/2005JB003726.

Machac, T.A., Zanner, C.W., Geiss1, Ch.E., 2007. Time dependent IRM acquisition as a tool to quantify the abundance of ultrafine superparamagnetic magnetite in loessic soils. Geophys. J. Int. 169, 483–489, doi: 10.1111/j.1365-246X.2007.03355.x.

Maher, B.A., 1998. Magnetic properties of modern soils and Quaternary loessic palaeosols: Palaeoclimatic implications. Palaeogeogr. Palaeoclimatol. Palaeoecol. 137, 25–54, doi:10.1016/S0031-0182(97)00103-X.

Maher, B.A., Thompson, R., 1991. Mineral magnetic record of the Chinese loess and paleosols. Geology 19, 3–6, doi:10.1130/0091-7613(1991) 019<0003:MMROTC> 2.3.CO;2.

Néel, L., 1949. Théorie du trainage magnétique des ferromagnétiques en grains fin avec application aux terres cuites. Ann. Geophys. 5, 99–136.

Neumann, J., 2006. Untersuchung von EM-Transienten einer Altlast auf superparamagnetischen Einfluss, Diplomarbeit, Universität zu Köln.

Neumann, J., Bergers, R., Helwig, S.L., Hanstein, T., Kozhevnikov, N., Tezkan, B., 2005. Messung der TEM-Antwort von Bodenproben, in: Ritter, O., Brasse, H. (Eds.), 21 Kolloquium Elektromagnetische Tiefenforschung Haus Wohldenberg, Holle, 3–7.10.2005, pp. 331–338.

Oldfield, F., Thompson, R., Dickson, D.P.E., 1981. Artificial enhancement of stream bedload: A hydrological application of superparamagnetism. Phys. Earth Planet. Inter. 26, 107–124, doi:10.1016/0031-9201(81)90103-5.

Pasion, L.R., Billings, S.D., Oldenburg, D.W., 2002. Evaluating the effects of magnetic soils on TEM measurements for UXO detection. Expanded Abstracts. Society of Exploration Geophysicists, Tulsa, OK, 1428–1431.

Pokorny, J., Suza, P., Pokorny, P., Chlupáèová, M., Hrouda, F., 2006. Widening power of low-field magnetic methods in the investigation of rocks and environmental materials using the Multi-Function Kappabridge Set. Geophys. Res. Abstr. 8, EGU 06-A-04141.

Pokorny, J., Pokorny, P., Suza, P., Hrouda, F., 2011. A multi-function Kappabridge for high precision measurement of the AMS and the variations of magnetic susceptibility with field, temperature and frequency, in: Petrovsky, E., Herrero-Bervera, E., Harinarayana, T., Ivers, D. (Eds.), The Earth's Magnetic Interior. IAGA Special Sopron Book Series, Springer, Vol. 1, pp. 292–301.

Sagnotti, L., Winkler, A., 2012. On the magnetic characterization and quantification of the superparamagnetic fraction of traffic-related urban airborne PM in Rome, Italy. Atmospheric Environment 59, 131–140.

Sekachov, M.Yu., Balashov, B.A., Sachenko, G.V., Vechkapov, O.V., Zakharkin, A.K., Tarlo, N.N., Mogilatov, V.S., Zlobinsky, A.V., 2006. The *Tsikl-7* System for TEM Surveys. Pribory i Sistemy Razvedochnoi Geofiziki, No. 1 (15), 44–46.

Squires, G.L., 1968. Practical Physics. Cambridge University Press, Cambridge.

Stognii, Vas.V., Kozhevnikov, N.O., Antonov, E.Yu., 2010. TEM surveys for magnetic viscosity of rocks *in situ*. Russian Geology and Geophysics (Geologiya i Geofizika) 51 (11), 1219–1227 (1565–1575).

Stoner, E.C., Wohlfarth, E.P., 1948. A mechanism of magnetic hysteresis in heterogeneous alloys. Philos. Trans. R. Soc. London, Ser. A, 240, 599–642, doi:10.1098/rsta.1948.0007.

Tabbagh, A., 1986. Application and advantages of the Slingram electromagnetic method for archaeological prospecting. Geophysics 51 (3), 576–584.

Tarduno, J.A., 1994. Temporal trends of magnetic dissolution in the pelagic realm: Gauging paleoproductivity? Earth Planet. Sci. Lett. 123, 39–48, doi:10.1016/0012-821X(94)90255-0.

Thiesson, J., Tabbagh, A., Flageul, S., 2007. TDEM magnetic viscosity prospecting using a Slingram coil configuration. Near Surface Geophysics, 363–374.

Thompson, R., Oldfield, F., 1986. Environmental Magnetism. Allen and Unwin, London.

Trukhin, V.I., 1973. Introduction into Rock Magnetism [in Russian]. Moscow University Press, Moscow.

Verosub, K.L., Roberts, A., 1995. Environmental magnetism: Past, present, and furture. J. Geophys. Res. 100, 2175–2192, doi:10.1029/94JB02713.

Wang, X., Løvlie, R., Zhao, X., Yang, Z., Jiang, F., Wang, S., 2010. Quantifying ultrafine pedogenic magnetic particles in Chinese loess by monitoring viscous decay of superparamagnetism. Geochemistry, Geophysics, Geosystems (G3) 11 (10), Q10008, doi:10.1029/2010GC003194

Worm, H.-U., 1999. Time-dependent IRM: A new technique for magnetic granulometry. Geophys. Res. Lett. 26 (16), 2557–2560, doi:10.1029/1999GL008360.

Worm, H.-U., Jackson, M., 1999. The superparamagnetism of Yucca Mountain Tuff. J. Geoph. Res. 104 (B11), 25,415–25,425.

Xie, Q., Chen, T., Xu, H., Chen, J., Ji, J., Lu, H., Wang, X., 2009. Quantification of the contribution of pedogenic magnetic minerals to magnetic susceptibility of loess and paleosols on Chinese Loess Plateau: Paleoclimatic implications. J. Geophys. Res. 114 (B09101), doi:10.1029/2008JB005968.

Yu, Y., Tauxe, L., 2006. Acquisition of viscous remanent magnetization. Phys. Earth Planet. Inter. 159, 32–42.

Zakharkin, A.K., Bubnov, V.M., 1995. A Geophysical Prospecting Tool. Invention Certificate 2045083 of the Russian Federation [in Russian]. Published in BI, 27.09.95, No. 27.

Zakharkin, A.K., Bubnov, V.M., Kryzhanovsky, V.A., Tarlo, N.N., 1988. Magnetic viscosity of rocks as a new complicating effect on TEM soundings, in: Rabinovich, B.I. (Ed.), TEM Surveys for Mineral Exploration in Siberia [in Russian]. Nauka, Novosibirsk, pp. 19–26.

Zhou, L.P., Oldfield, F., Wintle, A.G., Robinson, S.G., Wang, J.T., 1990. Partly pedogenic origin of magnetic variations in Chinese loess. Nature 346, 737–739, doi:10.1038/ 346737a0.

Editorial responsibility: A.D. Duchkov