SHORT COMMUNICATIONS

APPLICATION OF THERMOLUMINESCENCE THEORY TO THE INVESTIGATION OF THERMOREMANENT MAGNETIZATION CURVES

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Abstract

Because of the similarity that exists between the basic equations describing the behaviour of thermoluminescence (TL) and thermoremanent magnetization (TRM), it is shown here that previously established methods for analysis of TL glow curves can be usefully applied to the analysis of TRM curves.

The thermoremanent magnetization (TRM) and partial thermoremanent magnetization (PTRM) of synthetic and natural ilmenite–hematites have been investigated recently by Westcott and Parry (1968), Westcott-Lewis (1971), and Westcott-Lewis and Parry (1971). The theory of TRM and PTRM was previously established by Néel (1955) and Stacey (1963). As pointed out by Stacey (1961), the observed PTRM curve is not a sharp line at a particular temperature, even when only a single activation energy is involved, but is rather spread over a certain temperature range. According to Stacey (1963), the magnetic behaviour is governed by the differential equation‡

$$-\mathrm{d}M/\mathrm{d}t = CM\exp(-E/kT), \tag{1}$$

where t is the time in seconds, C is a frequency factor (s^{-1}), E is the activation energy (eV), k is Boltzmann's constant, and T is the absolute temperature. If we now assume a linear heating function

$$T = T_0 + \beta t, \tag{2}$$

the solution of equation (1) which may be obtained is in fact the TRM curve. Differentiation of this function with respect to T results in the PTRM curve. In practice, the PTRM curve is often derived by differentiating the observed TRM curve.

From the mathematical viewpoint, equation (1) is identical with the equation governing the behaviour of a first-order thermoluminescence (TL) glow peak. According to Randall and Wilkins (1945*a*, 1945*b*), the TL emitted by a crystal is given by the relation

$$I = -\mathrm{d}n/\mathrm{d}t = sn\exp(-E/kT), \tag{3}$$

where I is the TL intensity, n is the concentration of trapped electrons (or holes)

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‡ The right-hand side of equation (1) was multiplied by a factor of 3 in Stacey's paper. This assumes triaxial symmetry in the anisotropy of the materials, which we no longer have reason to believe.

(cm^{-3}), s is the frequency factor (s^{-1}), and E is the activation energy (eV) needed for the release of the electrons. The solution of this equation gives $n = n(T)$ for a given heating function. Under certain circumstances n represents a measurable colouring of the crystal (Balarin and Zetzsche 1962). The derivative $-dn/dt$, which is obtained when equation (3) is solved, represents the emission of the thermally stimulated luminescence. The explicit equation for I in this case is

$$I = n_0 s \exp(-E/kT) \exp\left(-(s/\beta) \int_{T_0}^{T} \exp(-E/kT')\, dT'\right), \tag{4}$$

where β is the constant heating rate and n_0 is the initial concentration of trapped carriers. The formula for the maximum is found by equating the derivative of (4) to zero, which yields

$$\beta E/kT_m^2 = s \exp(-E/kT_m), \tag{5}$$

where T_m is the temperature at which I reaches a maximum.

Several methods for the evaluation of the activation energy E from a glow curve have been developed. The most commonly used is the "initial rise" method (Garlick and Gibson 1948) which employs the TL intensities in the low temperature portion of the glow curve. This method is not expected to yield accurate results as far as the PTRM curves are concerned, since only very few experimental points can be obtained accurately in the low temperature region. Other methods for the determination of E are based upon the shape of the glow curve (e.g. Chen 1969). It is this type of calculation which we intend to apply to the PTRM curves.

A convenient formula for calculating the activation energy of a first-order glow peak (Chen 1969) is

$$E = 2{\cdot}52\, kT_m^2/\omega - 2kT_m, \tag{6}$$

where $\omega = T_2 - T_1$ and T_1 and T_2 are the temperatures at which the glow reaches values of half the maximum intensity below and above T_m respectively. The symmetry factor $\mu_g = \delta/\omega$, where $\delta = T_2 - T_m$, can be used for determining the order of the peak. Chen (1969) shows that $\mu_g = 0{\cdot}4$ is characteristic of first-order peaks while μ_g is slightly greater than $0{\cdot}5$ for second-order peaks (i.e. the peak is nearly symmetric).

We shall now use the methods of analysis developed above for TL peaks to analyse the PTRM peak given in Figure 3 of Westcott-Lewis and Parry (1971). This peak is readily seen to be asymmetric with $\mu_g < 0{\cdot}5$; thus the first-order behaviour is established in agreement with equation (1). T_m is seen to be 127°C and $\omega \approx 54$°C. Substitution into equation (6) results in the value $E = 0{\cdot}6$ eV. With this value of E in equation (5), and replacing s by C, we get $C/\beta \approx 1{\cdot}4 \times 10^6\ K^{-1}$.

The shape of a TL peak and its temperature of maximum intensity depend on the heating rate (see e.g. Chen and Winer 1970), and this also applies to the PTRM curves (Stacey 1961). Since in the TRM measurement, the data were obtained by heating the sample from room temperature to a temperature T and cooling back to room temperature, a relatively low effective heating rate is to be assumed, say $\beta = 0{\cdot}2\ K\,s^{-1}$, which yields $C = 3 \times 10^5\ s^{-1}$.

More precise results can be achieved by continuous recording of the TRM curve, so that the PTRM curve is established more accurately. The following improvement to the derived value of E can then be made. In TL theory, it is possible that s depends upon temperature according to the relationship

$$s \propto T^a,$$

for $-2 \leqslant a \leqslant 2$ (Chen 1969). The final term of equation (6) then has to be modified to $(2+a)kT_m$. According to Stacey (1963) C is proportional to T and so, for the PTRM case, the final term in equation (6) should be $3kT_m$. This reduces the result by some 5% or more.

The result of $E = 0 \cdot 6$ eV compares with the value of $0 \cdot 3$ eV found for similar oxides by Ishikawa and Syono (1963) and $1 \cdot 1$ eV by Westcott-Lewis (1972). Some of the other determinations of C yield values between 10^9 and 10^{13} s^{-1} (Stacey 1963), while more recently a value of $C \approx 10^8$ s^{-1} is considered to be acceptable.

In conclusion, methods previously established for analysis of TL glow curves have been shown to be valuable also for the analysis of PTRM curves. This follows directly because of the mathematical resemblance of the basic equations describing the TL and PTRM phenomena.

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