The rare-earth tellurates are a new class of inorganic phosphors with interesting luminescent properties. When activated by trivalent europium, they exhibit characteristics which are consistent with the observations made by numerous researchers in the field. The exact assignment of transitions is not possible pending the clarification and resolution of the crystal structure. The significant conclusion of the research with uranium activated tellurates is the indication that the identity of the molecular ion uranyl species is preserved in a solid crystalline matrix as demonstrated by its fluorescent properties. Further analysis is required to define more closely the luminescence mechanisms in uranium activated compounds.

Acknowledgment

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Glow Curves with General Order Kinetics

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ABSTRACT

Thermoluminescence and thermally stimulated current curves obeying general order kinetics laws are being investigated. For these cases, whose order is not necessarily first or second but rather may have some noninteger value, an effective method of calculating the activation energy is given. This method is based on measuring the temperature at the maximum of the glow peak and the half intensity temperatures. Numerical calculations for orders between 0.7 and 2.5, activation energies between 0.1 and 1.6 eV and frequency factors between 10^5 and 10^{13} sec^{-1} have been done using an I.B.M. 360 computer. The results reveal the general characteristics of these peaks and their dependence on the parameters. The new method for calculating the activation energy is also checked numerically.

The measuring of glow curves was found to be one of the most convenient ways to determine the activation energies of trapping levels in crystals. This includes the phenomena of thermoluminescence (TL) (1), thermally stimulated current (TSC) (2), thermally stimulated electron emission (TSEE) (3), and thermally stimulated capacitor discharge (TSCD) (4). The glow curves were analyzed usually by assuming first or second order kinetics. The first order case was investigated first by Randall and Wilkins (5) who assumed that the glow intensity I may be given by

$$I = - dn/dt = sn \, \exp\,(- E/kT) \qquad [1]$$

where n is the concentration of trapped carriers (cm^{-3}), t the time, s the frequency factor (sec^{-1}), sometimes referred to as "the preexponential factor," E is the activation energy (ev), k is Boltzmann's constant, and T the absolute temperature. By solving this differential equation and assuming a linear heating rate of β °K/sec, one has for the intensity

$$I = sn_o \, \exp(- E/kT)$$
$$\exp\left[- (s/\beta) \int_{T_o}^{T} \exp(- E/kT') \, dT' \right] \qquad [2]$$

where n_o is the initial concentration of carriers and T_o the initial temperature. The condition for the maximum of the peak is found by differentiating Eq. [2] and equating the derivative to zero. Thus

$$\beta E/(kT_m{}^2) = s \, \exp\,(- E/kT_m) \qquad [3]$$

where T_m is the temperature at the maximum. Garlick and Gibson (6) introduced the possibility of second order glow peaks, obeying the equation

$$I = - dn/dt = s'n^2 \, \exp\,(- E/kT) \qquad [4]$$

where s' is a "preexponential constant" (cm^3 sec^{-1}). The solution of the equation gives

$$I = s'n_o{}^2 \, \exp\,(- E/kT)$$
$$\left[1 + (s'n_o/\beta) \int_{T_o}^{T} \exp\,(- E/kT') \, dT' \right]^{-2} \qquad [5]$$

and the condition for the maximum is

$$1 + (s'n_o/\beta) \int_{T_o}^{T_m} \exp\,(- E/kT) \, dT$$
$$= \frac{2kT_m{}^2 \, s'n_o}{\beta E} \, \exp\,(- E/kT_m) \qquad [6]$$

Many methods for calculating the activation energies of glow curves have been given (2). Only one of these, the "initial rise" method (6) is expected to be useful for all the possible orders of the process. However, in many cases, the use of this method is limited because of experimental (7) and theoretical (8, 9) reasons.

A useful method of calculating the activation energy by using the temperature maximum T_m and the falloff half of the peak $\delta = T_2 - T_m$ where T_2 is the higher of the two half intensity temperatures, was given by Lushchik (10) for first and second order kinetics. Another method using the low temperature half width $\tau = T_m - T_1$ was developed by Halperin and Braner (11). These two methods were slightly modified by Chen (12) and are now more accurate and more easily usable. Chen also gave another method in which the total width $w = T_2 - T_1$ is used, and showed that in certain cases this method is preferable. The three methods were summed up (12) as

$$E_\alpha = c_\alpha (kT_m^2/\alpha) - b_\alpha (2kT_m) \qquad [7]$$

where α is δ, τ, or w. The values of c_α and b_α for the three methods and for first and second order processes are given in Table I. The equations [7] are slightly changed in cases where the preexponential factor depends on temperature as a power function $s = s'' T^a$ where usually $-2 \leq a \leq 2$ (11-13) (see below).

Halperin and Braner (11) showed that an easy way to determine the order of a peak is by checking the values of $\mu_g = n_m/n_o$ where n_m is the concentration of carriers at the maximum. Values of μ_g of about $(1 + \Delta)/e$ where $\Delta = 2kT_m/E$, should indicate first order kinetics, whereas values around $(1 + \Delta)/2$ indicate second order. As an aproximation to μ_g the value of $\mu_g' = \delta/w$ was taken. Chen (12) found that a characteristic value of μ_g' for first order peaks is 0.42 and for second order 0.52.

Although the conventional way for analyzing glow curves is assuming either first or second order kinetics, this by no means covers the general case even if only a single activation energy is involved. Halperin and Braner (11) wrote three simultaneous differential equations relating the variables $n =$ the concentration of trapped electrons, $m =$ the concentration of holes in centers, and $n_c =$ the concentration of free electrons. However, Halperin and Braner solved the problem only for those conditions leading to reduction of the three equations to the first or second order cases.

A better approximation for the general case may be given by the equation given by May and Partridge (14)

$$I = -dn/dt = s'n^l \exp(-E/kT) \qquad [8]$$

where l is not necessarily 1 or 2. The general case of $l \neq 1$ is essentially limited to samples in which the concentration of trapped carriers involved in the glow peak is equal to that of the empty centers. May and Partridge (14) proved that for simultaneous activation of two electrons, the value of l should be 1.5. This same value was found by them experimentally for TL peaks in KCL samples. Partridge and May (15) report the same order of 1.5 for NaCl samples after a certain heat treatment, whereas other peaks in NaCl yield values of 0.7 ± 0.1.

In the present paper, this problem of general order kinetics is further investigated. Some of the properties of the peaks are found by numerical calculations of the peaks' parameters by the use of an I.B.M. 360 computer. Several interpolation methods for cal-

culating the activation energies based on the methods of Lushchik, Halperin and Braner, and Chen for first and second order are introduced and checked. The method based on the measurement of τ is found to be the most accurate.

Theoretical Approach and Numerical Calculation

The solution of Eq. [8] gives for $l \neq 1$

$$I = s'n_o^l \exp(-E/kT) \left[\frac{(l-1)s'n_o^{(l-1)}}{\beta} \int_{T_o}^T \exp(-E/kT')dT' + 1 \right]^{-l/(l-1)} \qquad [9]$$

A special case of this equation for $l = 2$ is Eq. [5]. $s'n_o^{l-1}$ has the units of sec^{-1} and in this sense it is similar to s in the first order case. If one writes s instead of $s'n_o^{l-1}$, one has

$$I = sn_o \exp(-E/kT) \left[\frac{(l-1)s}{\beta} \int_{T_o}^T \exp(-E/kT')dT' + 1 \right]^{-l/(l-1)} \qquad [10]$$

Although Eq. [10] is not valid for the case $l = 1$, it can easily be shown that it reduces to Eq. [2] in the limit when $l \to 1$. It is in this sense that Eq. [10] is a general formula applicable for all possible values of l.

The condition for maximum is found by equating the derivative of [10] to zero

$$[(l-1) s/\beta] \int_{T_o}^{T_m} \exp(-E/kT) dT + 1$$

$$= \frac{slkT_m^2}{\beta E} \exp(-E/kT_m) \qquad [11]$$

Again, a special case for $l = 2$ would be Eq. [6]. It should also be noted that Eq. [11] reduces to Eq. [3] for $l = 1$ which might be expected from the fact that Eq. [9] and [10] are valid for the first order case in the sense of the limit for $l \to 1$.

In a way similar to what has been done previously (12) for first and second order peaks, Eq. [11] can be solved numerically for given values of l, s, E, and β (and assuming that T_m is sufficiently larger than T_o) to give the value of T_m. This is done by approximating the integral of the left hand side of Eq. [11] by a certain number of terms of the asymptotic series (7)

$$\int_{T_o}^T \exp(-E/kT')dT' \cong T \exp(-E/kT)$$

$$\sum_{n=1}^{\infty} \left(\frac{kT}{E} \right)^n (-1)^{n-1} n! \qquad [12]$$

and solving the equation by the iterative Newton-Raphson method (12).

This has been done for values of l between 0.7 and 2.5, values of s between $10^5 sec^{-1}$ and $10^{13} sec^{-1}$ and values of E between 0.1 and 1.6 eV. These ranges seem to cover practically all the parameters' values found experimentally. β was taken to be 0.5°K/sec and this parameter has not been varied since we can consider s/β as one parameter in Eq. [11] and s was varied by several orders of magnitude. As a first approximation for the iterative process, it was found useful to take $T_m = 500 \times E$. These calculated values of T_m are seen in column 4 of Table I for given values of E, s, and l shown in columns 1, 2, and 3, respectively. Only the cases with $1 < l < 2$ are given in some detail, whereas only examples for $l = 2.5$ and $l = 0.7$ are shown. The calculations were done, however, for the whole mentioned range and the conclusions are based on all of these results. It is to be noted that the computed values of T_m for fixed values of E and s depend

Table I. Coefficients appearing in Eq. [7] for the various methods of calculating activation energies

| | First order | | | Second order | | |
	τ	δ	w	τ	δ	w
c_α	1.51	0.976	2.52	1.81	1.71	3.54
b_α	1.58	0	1	2	0	1

very slightly on the value of l. This can be understood by examining Eq. [11]. By taking only the first two terms in the series [12] which is a good approximation for most of the cases (11), Eq. [11] yields

$$\beta E/(kT_m{}^2) = s \exp(-E/kT_m)[1 + (l-1)\Delta] \quad [13]$$

where $\Delta = 2kT_m/E$. This again, reduces to the simple equation [3] for the first order case ($l = 1$). Since Δ is usually of the order of magnitude of 0.1, the term in brackets changes by 20% at most for extreme l values, which may cause changes of only around 1% in T_m because of the appearance of T_m in the exponential. Thus we may expect T_m to depend only very slightly on l for the same values of E and s, which is clearly seen in column 4.

Once the value of T_m is found, the intensity at the maximum I_m can be found by inserting T_m into Eq. [9] and using again the asymptotic series as a good approximation for the integral.

Now the values of T_1 and T_2, the low and high temperatures of half intensity, can be calculated by solving numerically the equation $I(T) = I_m/2$, when $I(T)$ is given by Eq. [9]. Again, the series approximating the integral is used throughout the iteration process. As a convenient first approximation we take $0.95\ T_m$ for T_1 and $1.05\ T_m$ for T_2. A similar process with somewhat more details was given previously by Chen (12) for the case of $l = 1$ and $l = 2$. By using the T_1, T_m, and T_2 values, the parameters $\delta = T_2 - T_m$ (the high temperature half width), $\tau = T_m - T_1$ (the low temperature half width and $w = T_2 - T_1$ (the total half width) are easily found, and shown in columns 5, 6, and 7 of Table I, respectively. The geometrical factor $\mu_g' = \delta/w$ is found and shown in column 8. Figure 1 gives calculated values of μ_g' as a function of the given l values. The upper and lower curves give the limits of variations of μ_g' values when E and s are varied, whereas the center curve shows the average values. This curve may be used for estimating the value of l by the measured μ_g's. The possible error is seen not to exceed $\pm 7\%$. The curve gives more information than column 8 in Table II since the given average values and possible deviations (for various E and s values) are based on many sets of calculations not included in the table. Another factor characterizing the geometrical shape of a peak, namely, $\gamma = \delta/\tau$ (13) can be directly found, this is shown in column 12.

Methods for Calculating E and s

In order to find the activation energies by one of the half width methods, it is suggested to interpolate (and to some extent extrapolate) the constants appearing in the first and second order equations [7]. The interpolation could be done according to the values of l. However, this magnitude is not found directly by the experimental results. A much more convenient interpolation parameter seems to be μ_g' which is found directly and easily from the geometrical shape of the peak. μ_g' depends almost only on l, with only minor dependence on E and s (Fig. 1). Thus, interpolation with respect to the argument μ_g' is in a way interpolation with respect to l. We tried first, naturally, the simplest way of interpolation, namely, the linear interpolation. We have to write the general equations so that they would give the first order case for $\mu_g' = 0.42$ and the second order one for $\mu_g' = 0.52$ (12). With the coefficients given in Table I, the factors in Eq. [7] for the interpolated-extrapolated τ method would be

$$c_\tau = 1.51 + 3.0\ (\mu_g' - 0.42); \quad b_\tau = 1.58 + 4.2\ (\mu_g' - 0.42) \quad [14]$$

For the equation using the δ value one has

$$c_\delta = 0.976 + 7.3\ (\mu_g' - 0.42); \quad b_\delta = 0 \quad [15]$$

and for the w method we find

$$c_w = 2.52 + 10.2\ (\mu_g' - 0.42); \quad b_w = 1 \quad [16]$$

Table II. Calculated parameters for given energies, frequency factors, and orders of kinetics

1 E	2 s	3 l	4 T_m	5 δ	6 τ	7 w	8 μ_g'	9 E_δ	10 E_τ	11 E_w	12 γ
1.6	10^{13}	2.5	554.1	33.9	28.1	62.0	0.548	1.486	1.583	1.535	1.210
0.1	10^5	2.5	82.1	11.4	8.8	20.2	0.566	0.104	0.098	0.101	1.302
1.6	10^{13}	1.9	554.6	27.4	26.4	53.8	0.509	1.575	1.599	1.595	1.038
0.4	10^{13}	1.9	144.3	7.4	7.1	14.5	0.510	0.395	0.400	0.399	1.041
0.1	10^{13}	1.9	37.6	2.0	1.9	3.9	0.511	0.099	0.100	0.100	1.043
1.6	10^9	1.9	746.3	49.2	46.5	95.7	0.514	1.623	1.597	1.618	1.060
0.4	10^9	1.9	196.7	13.7	12.8	26.5	0.516	0.408	0.399	0.408	1.064
0.1	10^9	1.9	52.0	3.8	3.6	7.4	0.517	0.103	0.100	0.102	1.069
1.6	10^5	1.9	1124.3	110.3	100.2	210.5	0.524	1.713	1.589	1.659	1.100
0.4	10^5	1.9	303.6	32.1	28.9	61.0	0.526	0.434	0.397	0.417	1.110
0.1	10^5	1.9	82.5	9.4	8.4	17.8	0.529	0.110	0.099	0.105	1.121
1.6	10^{13}	1.5	554.9	22.7	25.0	47.7	0.476	1.616	1.606	1.623	0.907
0.4	10^{13}	1.5	144.4	6.1	6.7	12.9	0.476	0.406	0.402	0.406	0.910
0.1	10^{13}	1.5	37.6	1.7	1.8	3.5	0.477	0.102	0.100	0.102	0.912
1.6	10^9	1.5	747.1	40.9	44.2	85.1	0.481	1.668	1.605	1.645	0.926
0.4	10^9	1.5	196.9	11.4	12.2	23.6	0.482	0.420	0.401	0.412	0.930
0.1	10^9	1.5	52.0	3.2	3.4	6.6	0.483	0.106	0.100	0.103	0.934
1.6	10^5	1.5	1126.6	92.3	95.9	188.2	0.490	1.765	1.598	1.687	0.962
0.4	10^5	1.5	304.3	26.9	27.7	54.6	0.492	0.447	0.399	0.424	0.970
0.1	10^5	1.5	82.7	7.9	8.1	16.0	0.495	0.113	0.100	0.107	0.980
1.6	10^{13}	1.1	555.2	16.7	23.3	40.9	0.431	1.591	1.605	1.613	0.757
0.4	10^{13}	1.1	144.5	4.8	6.3	11.1	0.431	0.399	0.401	0.404	0.759
0.1	10^{13}	1.1	37.6	1.3	1.7	3.0	0.432	0.100	0.100	0.101	0.761
1.6	10^9	1.1	747.8	31.9	41.3	73.2	0.436	1.647	1.604	1.635	0.772
0.4	10^9	1.1	197.1	8.7	11.4	20.3	0.437	0.414	0.400	0.409	0.775
0.1	10^9	1.1	52.1	2.5	3.2	5.7	0.438	0.104	0.100	0.103	0.778
1.6	10^5	1.1	1129.1	72.4	90.4	162.7	0.445	1.755	1.598	1.676	0.801
0.4	10^5	1.1	305.1	21.1	26.1	47.3	0.447	0.445	0.399	0.421	0.808
0.1	10^5	1.1	82.9	6.2	7.6	13.9	0.449	0.113	0.100	0.106	0.816
1.6	10^5	0.7	1131.6	49.7	82.6	132.2	0.376	1.449	1.568	1.530	0.602
0.1	10^8	0.7	57.6	2.1	3.5	5.6	0.372	0.086	0.098	0.094	0.591

The results found by [14], [15], and [16] calculated by the computed μ_g' values are given in columns 9, 10, and 11 of Table II, respectively. These results may be compared to the known energies in order to evaluate the accuracy of each method separately. It is seen very clearly that the methods using the values of δ or w give poor evaluations for the activation energy, whereas Eq. [14] gives results which are always within 2% or better of the correct E value. This is true not only for the examples given in Table II but also for all the checked possibilities (see the ranges of the parameters above). Thus we cover most of the experimentally possible cases.

Once the activation energy is known, an estimate on the frequency factors can be found by the use of Eq. [13]. A reasonable value of s is found even when the factor $(l - 1)\Delta$ is neglected. A better evaluation can be calculated by estimating the value of l according to the measured μ_g' and using Fig. 1. It is self evident
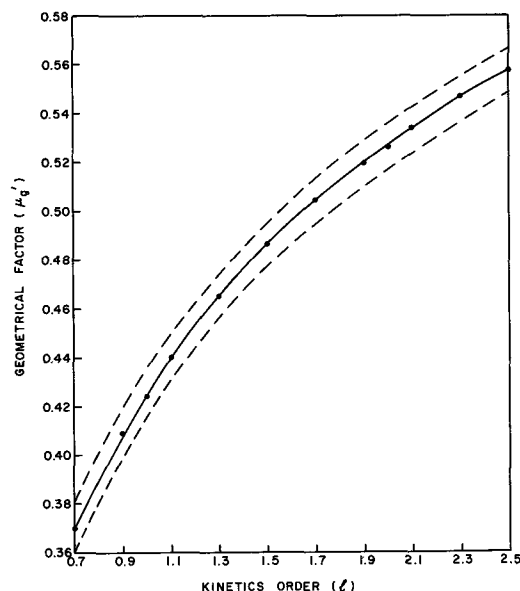

Fig 1. Calculated geometrical factor (μ_g') as a function of the given kinetics order (*l*). Central line gives the average values, upper and lower lines give the largest possible variations (for various E's and s's) for the given *l*'s.

that the better the approximation in finding E, the better the corresponding calculated factor s. In order to find the constant $s'(s = s'n_o{}^{l-1})$, we have to have some additional information about n_o.

Discussion

Glow curves obeying a general kinetic law of the form given by Eq. [8] have been investigated. A method for calculating activation energies by the use of the maximum temperature and the low temperature half width was found by linear interpolation between the known formulas of first and second order kinetics. Similar methods using the high temperature half width and the total half width have failed. It is possible that by the use of nonlinear interpolation, better formulas could be found for the energy. However, this would make the equations more complicated. The results found by Eq. [14] are accurate to about 2%, which is comparable to the precision of the original first and second order formulas. This seems to be satisfactory for practically all the cases, especially since the possible experimental errors may contribute higher inaccuracies. Moreover, it has been shown (11, 12) that methods using the low temperatures half width are better in the sense that this portion of the peak can be "cleaned" by thermal bleaching in order to get rid of possible satellites, which would result in better calculated energy values.

It should be mentioned that the success of interpolating the equations using the low temperatures half width seems to be due to the fact that the value of τ does not depend strongly on l. The error done by using the first order τ equation for a second order peak, for example, would not exceed usually 20%. The reduction of the possible error to less than 2% by interpolation seems, however, to be essential.

Interpolation with respect to the other geometrical factor γ (7) in the same way yielded results about as good as the mentioned ones when the τ method was used, and about as bad for the δ and w methods. Another method employing the value of τ for finding the activation energy was given by Grosswiener (16) as follows

$$E_G = a_\tau kT_1T_m/\tau \qquad [17]$$

where $a_\tau = 1.41$ for first order and $a_\tau = 1.68$ for second order (12). By interpolation between these two coefficients according to the μ_g' values, results good to about 2% were found again.

The present method for estimating the value of l by the use of Fig. 1 seems to be quite reliable and appreciably simpler than the method mentioned by May and Partridge (14). An important point to be emphasized is that the present investigation gives a better insight to the relation between the various order kinetics. Equations [8], [9], [10], [11], and [13] are applicable for all values of l including 1 and 2 (Eq. [9] and [10] in the sense of the limit for $l \to 1$). Although the present treatment does not cover the problem of general glow peak associated with one activation energy, this approach seems to give much more reliable results for the calculated parameters than just using first or second order assumptions, which is the conventional approach.

Finally, it has to be noted that we dealt here only with the case of frequency factors independent of temperature. It has been mentioned above that in some cases s depends on temperature as some power function, $s = s'' T^a$ where s'' is a constant and $-2 \le a \le 2$. For this case, it has been shown (12) for first and second order kinetics that a better approximation can be found by subtracting akT_m from the value calculated by any of the usable methods. It can be shown that this correction is applicable for the presently discussed more general case, and thus, when "a" is known by some other measurements, it may be used. This correction seems to be significant, since in some cases it may change the calculated energy value by up to 10%.

Symbols

E	activation energy, eV
s	frequency factor, $\sec^{-1}$
s'	preexponential factor, $\sec^{-1} cm^{3(l-1)}$
l	kinetics order
T, T'	temperatures, °K
T_o	initial temperatures, °K
T_m	temperature at the maximum, °K
T_1, T_2	half intensity temperatures, °K
β	heating rate, °K/sec
t	time, sec
k	Boltzmann constant, eV/°K
I	glow intensity
I_m	maximal glow intensity
n	concentration of trapped electrons, cm^{-3}
n_o	initial concentration of trapped electrons, cm^{-3}
n_m	concentration of trapped electrons at the maximum, cm^{-3}
m	concentration of holes in centers, cm^{-3}
n_c	concentration of free electrons, cm^{-3}
τ	low temperature half width, °K
δ	high temperature half width, °K
w	total half width, °K
$E_\alpha, E_\tau, E_\delta, E_w$	activation energies calculated by various methods, eV
$c_\alpha, b_\alpha, c_\tau, b_\tau, c_\delta, b_\delta, c_w, b_w, a_\tau$	constants appearing in various equations for finding activation energies
Δ	correction factor $(=2kT_m/E)$
μ_g	characteristic factor $(=n_m/n_o)$
μ_g'	geometrical factor $(=\delta/w)$
λ	geometrical factor $(=\delta/\tau)$

Any discussion of this paper will appear in a Discussion Section to be published in the June 1970 JOURNAL.

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