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# Thermoluminescence and activation energies in $Al_2O_3$, MgO and LiF (TLD-100)

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Thermoluminescent characteristics of $Al_2O_3$ and MgO single crystal samples obtained from several sources have been determined after X-ray and ultra-violet irradiation. The glow peak spectrum above room temperature, the emission spectrum, the impurity content and distribution and the activation parameters for the observed glow peaks are reported. Czochralski-grown $Al_2O_3$ samples indicate a major glow peak, the position of which shifts to lower temperatures with increasing dose, an observation with interesting implications in thermoluminescent radiation dosimetry. MgO also has characteristics which could be useful in dosimetry, especially in the ultra-violet region. Activation parameter comparisons are made for the observed glow peaks in $Al_2O_3$ and MgO and in LiF (TLD-100). After considering a number of theoretical and experimental problems and uncertainties, it is concluded that determinations of activation parameters are less meaningful than observations of other thermoluminescent characteristics in the understanding of thermoluminescent behaviour.

## 1. Introduction

Thermoluminescence (TL) has long been used as an experimental method in the study of electron and hole traps in solids, as well as in the study of kinetics of trapping and recombination processes. More recently, radiation dosimeters utilizing the TL output of certain materials have been developed for application in the detection of ionizing radiation. The need for dosimeter materials with high sensitivity, reproducibility and with a linear dose response has emphasized the need for a basic understanding of the TL process, which has so far been lacking.

An example of a material widely used for X- and gamma-ray dosimetry is LiF (TLD-100) (Harshaw Chemical Co), which has relatively well-characterized TL characteristics [1, 2]. $Al_2O_3$ and MgO are also potentially useful dosimeter materials in the X-ray and the ultra-violet regions of the spectrum, as is $CaF_2$. In all of these materials, the TL properties are dependent upon impurities and impurity complexes, the identities of which are generally unknown except in LiF where the influence of impurities is somewhat better understood [3, 4].

In the evaluation and comparison of TL data in the literature, several problems become apparent. Most experimental samples are produced by different means and under different circumstances so that they have varying impurity species and concentrations as well as different impurity distributions. In addition, the reported TL peak temperatures are difficult to compare due to different heating rates and varying thermal contacts. Glow peaks are shifted to higher temperatures as the heating rate is increased, resulting in an apparent change in glow peak shape if the luminescent efficiency varies with temperature. Thus, if several peaks occur in the same temperature region, their identity may not be certain, especially if only peak temperatures are given without reference to heating rate. In addition, Dussel and Bube [5] point out that the temperature of the maximum is affected by the density of deep traps, so that the same peak in different crystals may appear at slightly different temperatures.

The kinetics of the TL process are usually reported in terms of a linear decay rate $W(T)$ assumed to be of the form $W(T) = s \exp(-E/kT)$ where $s$ is an "escape frequency factor" and $E$ is the "activation energy" for escape from

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*Present address: 4406 Amazon Drive West, Eugene, Oregon 97405, USA the trap involved. In the past, the comparison of values of $E$ and $s$ for different peaks has enabled the comparison of the influence of impurities, radiation damage and other variables on the TL process. However, recent theoretical analyses of the simple kinetic model of TL by Kelly and Braunlich [6] and by Saunders [7] have indicated the necessity of a more complete determination of trapping parameters than is available from the analysis of TL glow curves using simple models.

The purpose of the present paper is to investigate the TL output of several materials to enable an evaluation of their eventual usefulness as dosimetric materials. In this process, experimental problems related to temperature measurement and the usefulness of "activation energies" for comparisons between materials are also explored. The present study is concerned with TL in $Al_2O_3$ and MgO, with the use of additional data for LiF (TLD-100) for comparison purposes.

## 2. Experimental considerations

The measurement of the temperature of the luminescing crystal is critical and non-trivial in a TL experiment. This is generally due to the necessity of good thermal contact between the heater and the crystal and is also effected by poor thermal conduction within many samples (this is especially acute in powdered samples). In the present experiments, the most satisfactory results have been obtained using thin crystals, of approximately 4 × 4 × 1 mm, which were glued to the heater using VAC-SEAL, a high vacuum sealant (Environmental Space Sciences, Inc). This sealant provided a uniform, high conductivity contact surface for thermal conduction between the sample and the heater. Without the film of sealant between the heater and the sample, an apparent shift in glow peak temperature of 10 to 15°C was found. The sample chamber was evacuated to reduce convection cooling of the sample face. After an experiment, the sample could be removed by heating to about 150°C to soften the sealant.

The chromel-alumel thermocouple used to monitor the heater temperature was soldered to the heater directly behind the sample position. The true sample temperature was determined using this thermocouple by referring to a calibration plot of heater thermocouple emf versus the emf of a thermocouple embedded in a hole in a test crystal mounted in the usual sample position. To obtain a true reading during the calibration runs on the test crystal, it was found that the hole for the thermocouple in the test crystal should also contain some of the liquid high vacuum sealant discussed above. Under these circumstances, the sample was found to be 4°C lower than the heater at 200°C. If none of the sealant was used in the test crystal, the apparent sample temperature was 25°C below that of the heater at 200°C.

This illustrates one of the experimental difficulties which may be responsible for the experimental discrepancies between reported peak temperatures observed in the literature. During the analysis of activation parameters for glow curves, discussed below, it was found that an error of one degree in peak half-width could change the value of the activation energy by as much as 7 %. This emphasizes the importance of accurate temperature measurements in TL experiments.

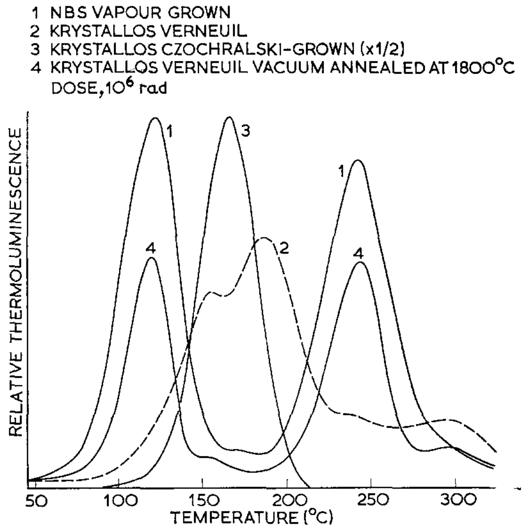

*Figure 1* X-ray induced thermoluminescence of aluminium oxide single crystals, as noted (Dose $10^6$ rad). Intensity of curve 3 reduced by one half. Heating rate 20°C/min.

## 3. Thermoluminescent characteristics

### 3.1. Aluminium oxide

Typical X-ray induced TL curves for several types of $Al_2O_3$ crystals after $10^6$ rad exposure are shown in Fig. 1. The samples obtained from the National Bureau of Standards were grown by vapour transport from ultrapure starting material [8]; growth was at 1740°C and the crystals were cooled to room temperature at 3°C/min. The Verneuil crystals were supplied by Krystallos, Inc, while the Czochralski-grown crystals were manufactured by Krystallos from similar Verneuil starting material. Spectrographic analysis of the Verneuil samples indicate that they are quite impure, especially in terms of iron and silicon impurities, which are present to several hundred ppm. The Czochralski-grown samples probably have a slightly lower impurity content and should be closer to equilibrium due to the slower growth process. The NBS samples should also be close to equilibrium, and are of a very high purity, with typical total impurity content less than 1 ppm.

In the as-received condition, the high purity NBS crystal shows a typical two-peak glow-curve pattern, with peaks near 120 and 240°C, as shown by curve 2 in Fig. 1. The as-received Verneuil samples (curve 2), on the other hand, show a broad TL curve with maxima near 155 and 185°C. The latter is similar to the TL behaviour reported by Cooke and Sutherland [9] in X-irradiated $Al_2O_3$ supplied by the Linde Company.

The as-received Krystallos Czochralski-grown samples give the single X-ray induced TL peak shown at 160°C (curve 3, Fig. 1). This glow peak has the interesting characteristic that its peak position shifts to lower temperatures with increasing exposure. This is true in all the Krystallos Czochralski-grown samples supplied to us. The peak position could be saturated at about $10^5$ rad, beyond which the peak temperature did not shift. The peak temperature shifts from 170°C at an exposure of 100 rad to 160°C at an exposure of $10^5$ rad. Changes in the glow peak shape with changing exposure were minor. Other authors (Van Tright and Van Der Kraay [10], Gabrysh *et al* [11]) apparently have not observed the shift in glow peak temperature with dose, perhaps due to the difficulties encountered with several overlapping peaks in their samples. This peak has interesting dosimetric possibilities, as discussed later.

Further experiments were performed using these $Al_2O_3$ crystals after annealing at 1800°C for 3 h in vacuum, followed by cooling at 2°C/min to room temperature. Under these conditions, the TL curve of the Verneuil crystals separate into a two-peak pattern which is strikingly similar to that obtained from the as-received NBS sample, as shown by curve 4, Fig. 1. The TL curve of the Krystallos Czochralski-grown samples also separates into two peaks, similar to the behaviour of the Verneuil samples. The TL output of these annealed samples is not identical to that of the NBS sample, however, since the emission spectrum of the Verneuil samples show a red emission only, while the NBS sample shows both red and blue emissions.

Buckman *et al* [12] have shown that certain types of $Al_2O_3$ are sensitive to ultra-violet irradiation. In the present work, neither the NBS nor the Verneuil samples showed detectable ultra-violet sensitivity. The Krystallos Czochralski-grown sample did, however, show ultra-violet sensitivity and gave a glow curve similar to that observed by Buckman *et al* [12] for crystals obtained from Semi-elements, Inc.

### 3.2. Magnesium oxide

Typical TL glow curves for three MgO samples under X-ray and ultra-violet irradiation are shown in Figs. 2 and 3. MgO samples Spicer I and Spicer Fe were obtained from the W. & C. Spicer Company as high purity and 300 ppm iron-doped samples, respectively. Sample Mono I was obtained from the Monocrystals Company. The high purity MgO crystals obtained from Spicer are generally the purest available (see also Srinivasan and Stoebe [13]). Spectrographic analysis shows Ca and Fe as major impurities in both Spicer I and Mono I crystals, with Ca levels of 100 and 200 ppm by weight, respectively, and Fe levels of 30 and 60 ppm. The Fe concentration in Spicer Fe was determined to be 300 ppm, with the Ca concentration at 1000 ppm.

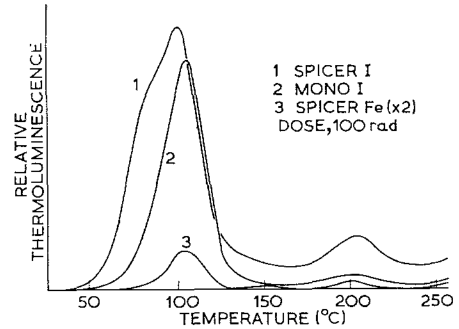

*Figure 2* X-ray induced thermoluminescence of magnesium oxide single crystals, as noted (Dose 100 rad). Intensity of curve 3 increased by a factor of two. Heating rate 20°C/min.

The glow curves obtained after X-irradiation in Fig. 2, and after ultra-violet irradiation in Fig. 3, show two main peaks. The peak near 100°C is the dominant peak present in most TL investigations in MgO. The intensity of this peak under both X-ray and ultra-violet irradiation is highest in sample Spicer I, decreasing in the other samples, the decrease apparently being proportional to the increasing concentrations of Fe and Ca impurities. The glow curves shown in Fig. 3 after ultra-violet irradiation at 313 nm show the presence of other lower temperature peaks in the Mono I and Spicer Fe samples (curves 2 and 3). In the Spicer Fe crystal, the dominant peak near 100°C is nearly absent after irradiation at 313 nm, but is greatly enhanced by irradiation at 250 nm, as shown by curve 4. Another low temperature peak appears in the Spicer I crystals in the X-ray case (curve 1, Fig. 2).

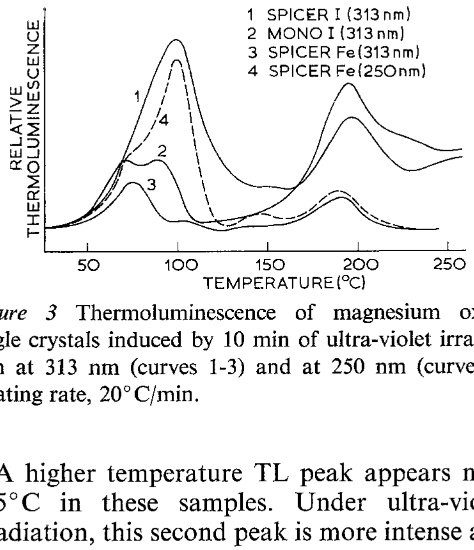

*Figure 3* Thermoluminescence of magnesium oxide single crystals induced by 10 min of ultra-violet irradiation at 313 nm (curves 1-3) and at 250 nm (curve 4). Heating rate, 20°C/min.

A higher temperature TL peak appears near 195°C in these samples. Under ultra-violet irradiation, this second peak is more intense and shows a somewhat different impurity dependence from that of the 100°C peak, as seen in Fig. 3. This higher temperature peak would be much more satisfactory for dosimetric applications than the dominant peak, since it should be much less sensitive to thermal fading.

Most of the literature concerning TL in MgO has concentrated on the identification of the emitting and trapping centres [14] and with the correlation of TL with changes in optical absorption bands [15] and with changes in impurity valence states [16]. Results from this laboratory for some of these other aspects of TL in MgO, and a discussion of the wavelength dependence under ultra-violet irradiation and of impurity effects, will be reported elsewhere [17].

## 4. Activation parameters

### 4.1. General considerations

The basic kinetic equations governing TL have been developed by a number of authors over the years. Methods of analysis in terms of the "activation energy" and "escape frequency factor," $E$ and $s$, generally have been based on the equations of Randall and Wilkins [18] and the further developments of Garlick and Gibson [19] which have recently been further analysed by Moran *et al* [20, 21]. The analyses of Saunders [7] and Kelly and Braunlich [6] based on general kinetic equations, have shown that glow-curve shapes depend upon parameters such as the ratio of retrapping to recombination rates, the number of thermally activated traps, the number of deep traps, and trap occupancy, as well as experimental factors such as the initial temperature and heating rate. Since these parameters are generally unavailable from analyses of glow peaks alone, the determination of activation energies and frequency factors from glow peaks cannot be strictly valid. However, the determination of such parameters could be useful for the comparison of experimentally observed glow peaks provided that experimentally determined values of these activation parameters are consistent with one another. This postulate is investigated in the present work.

The most satisfactory method for the analysis of TL activation parameters seems to have been the "initial rise" method [19]. This method is applicable to a wide range of kinetic conditions [6, 7], although some theoretical and experimental reasons limit its use [7, 23]. Alternate techniques for the determination of activation parameters are generally based on the shape of the glow peak [22, 23], and involve relationships between the activation energy, the peak temperature, and the temperature at half maximum on either (or both) side of the peak. Since peak-shape analyses are simple and straightforward, they have been popular with experimental workers, but few studies are available in which different methods for the determination of activation parameters are compared.

In an attempt to evaluate the usefulness of the various experimental methods available for activation parameter determinations, and the significance of these parameters themselves, values of $E$ and $s$ were determined in $Al_2O_3$ and

MgO and compared with values in the literature. Since activation parameter values available in the literature in these systems were generally determined using the initial rise or related methods, experiments were performed using peak-shape analyses only. The results for certain peaks in each material are discussed separately below.

### 4.2. Aluminium oxide

Values for the activation energy and frequency factor for the single peak in Krystallos Czochralski-grown $Al_2O_3$ are given in Table I in comparison with other values in the literature. Values reported for the present study use the method of Kelly and Laubitz [23] applied to the low temperature half-width of the peak obtained upon heating at a quadratic heating rate. Values are calculated for both first and second order processes; here, the second order calculation compares most favourably with the other results reported in Table I.

The activation parameter values obtained for the single peak in the present study and those for the low temperature peaks in the literature are seen to agree relatively well with one another, independent of the method used. During the peak shift to lower temperatures, which is observed with increasing irradiation, the calculated activation energy increases approximately 5% with dose compared to the values reported in Table I.

### 4.3. Magnesium oxide

Activation parameters for TL peaks in four MgO crystals are given in Table II. Values in the present work were obtained after ultraviolet irradiation at 313 nm for samples Spicer I and Mono I, and at 250 nm for Spicer Fe. Values obtained using both first and second order Kelly-Laubitz equations have been shown for one peak. Here, it is noted that the first order calculations appears to be more valid since the values obtained from the second order calculations are quite large.

Data for the dominant peak in MgO available in the literature is presented in Table II for comparison with the present results. Searle and Glass [24] applied the initial rise method to this peak, and using the correlated decay of the $V_1$ band, assert that the process is second order. This observation is in disagreement with the apparent validity of a first order process observed in the present work. However, the agreement among authors is generally good when considered in the context of the theoretical and experimental problems discussed earlier.

## 5. Discussion

The results given above indicate several characteristics of $Al_2O_3$ and MgO which may be useful in dosimetric applications. Samples of both materials can be obtained that are sensitive to either X-ray or ultra-violet irradiation or both, and they both exhibit glow peaks at temperatures high enough to avoid fading problems due to room temperature storage.

A decrease in glow peak temperature with increasing exposure, along with second order kinetics for the TL process, is predicted by Kelly and Braunlich [6] and by Saunders [7] under strong retrapping conditions. This fits in well with the experimental results for the major peak in the Krystallos Czochralski-grown $Al_2O_3$. A similar decrease in peak temperature with increasing exposure has been reported for one TL peak in $ThO_2$ by Rodine and Land [25], which may also be attributed to strong

TABLE I Activation energies and frequency factors for TL peaks in $Al_2O_3$ crystals after X-irradiation.

| Reference | Peak temperature | Heating rate | Method | Activation energy (eV) | Log frequency factor |
|---|---|---|---|---|---|
| Van Tright and Van Der Kraay [10] | 139 | ? | Isothermal decay from Randall-Wilkins [18] | 1.22 | 14.3 |
| | 168 | | | 1.31 | 13.6 |
| | 221 | | | 1.65 | 15.4 |
| Gabrysh *et al* [11] | 146 | Linear 50° C/min | (a) Grosswiener [29] | $1.15^a$, $0.95^b$ | — |
| | 180 | | (b) Luschik [30] | $1.27^a$, $1.11^b$ | — |
| | 227 | | equations | $1.56^a$, $1.20^b$ | — |
| Present study | 160 (saturated peak) | Quadratic $1.3 \times 10^{-6}$ (°C sec)$^{-1}$ | Kelly-Laubitz [23] | | |
| | | | First order | 1.08 | 11 |
| | | | Second order | 1.30 | 13 |

TABLE II Activation energies and frequency factors for prominent TL peaks in MgO. The peak denoted by (D) is the "dominant" peak seen in all MgO samples.

| Reference | Peak temperature (°C) | Heating rate | Method | Activation energy (eV) | Log frequency factor |
|---|---|---|---|---|---|
| Yamaka [31] | 77 (D) | Linear | First order | 0.99 | 12 |
| | 180 | 5°C/min | Randall-Wilkins [18] | 1.29 | 12 |
| Hecht and Taylor [32] | 82 (D) | Linear 2.7°C/min | First order Randall-Wilkins [18] | 0.926 | 13 |
| Searle and Glass [24] | 78 (D) | Linear 20°C/min | Initial rise Garlick and Gibson [19] | 1.12 | 15 |
| Wertz *et al* [14] | 86 (D) | Quadratic $5 \times 10^{-7}$ $(°C\ sec)^{-1}$ | First order Kelly-Laubitz [23] | 0.85, 0.90 | — |
| Present study | | Quadratic $1.3 \times 10^{-6}$ $(°C\ sec)^{-1}$ | Kelly-Laubitz [23] | | |
| Spicer Fe | 97 (D) | | First order | 1.13 | 14 |
| | 97 (D) | | Second order | 1.36 | 16 |
| | 191 | | First order | 1.40 | 13 |
| Spicer I | 96 (D) | | First order | 1.03 | 12 |
| | 68 | | First order | 0.97 | 12 |
| Mono I | 88 (D?) | | First order | 1.22 | 15 |
| | 197 | | First order | 1.33 | 12 |

retrapping. Another of the TL peaks in $ThO_2$ shifts the opposite direction (i.e. to higher temperatures with higher dose) probably due to the presence of two near-by peaks with different exposure response. The presence of two near-by peaks could also be the case with the observed shifting peak in $Al_2O_3$; additional work would be required to determine precisely the cause of the exposure-dependent peak position.

It is interesting to consider the dosimetric implications of the peaks in $Al_2O_3$ and $ThO_2$ which shift with dose. The current use of dosimeters based on TL requires a measurement of luminescent intensity in order to determine the dose received by the dosimeter crystal. In principle, a simpler and more easily calibrated measurement of peak temperature would, with prior tabulation of peak temperature versus dose, yield the same result. An important advantage of such a method would be that neither the amount of material used nor the energy of the radiation would affect the measurement. The mounting of the sample would, of course, be critical for accurate temperature measurement, but the method described in the present paper is both rapid and generally reproducible. The use of such a method would not depend upon the cause of the dose-dependent peak position, but only upon the amount of peak shift per unit dose. Unfortunately, for the shifting peaks in both $Al_2O_3$ and in $ThO_2$, the observed temperature shift is probably too small for use in such a dosimetry system.

The activation parameter results given for $Al_2O_3$ and MgO in Tables I and II, illustrate the problems of reproducibility in peak temperature and in values of the activation parameters themselves. However, values for corresponding peaks generally seem to agree, certainly to within 20%, with values determined by others using different techniques. In most cases, problems associated with thermal contact and temperature measurement could be much more important sources of uncertainty than are the theoretical problems.

In order to further determine the validity of activation parameter determinations, additional experiments have been performed using LiF (TLD-100). Because it is quite uniform in its TL behaviour, the results of various authors reporting glow-peak temperatures and activation energies should be more directly comparable than in other materials. The glow-peak structure is well known and the glow peaks above room temperature are generally numbered from

TABLE III Activation energies and frequency factors for certain TL peaks in Lif:TLD-100 obtained using X-irradiation.

| Reference | Peak | Peak temperature (K) | Heating rate | Method | Activation energy (eV) | Log-frequency factor |
|---|---|---|---|---|---|---|
| Zimmerman *et al* [26] | 4 | — | — | Isothermal decay from Randall-Wilkins [18] | 1.19 ± 0.5 | 11–12 |
| | 5 | — | | | 1.25 ± 0.6 | 11.12 |
| | 6 | — | | | 2.1 | 22 |
| Jackson and Harris [27] | 2 | 115 | — | Isothermal decay from Randall-Wilkins [18] | 0.84 ± 0.05 | 8–10 |
| | 3 | 160 | | | 0.89 ± 0.09 | 8–10 |
| | 4 | 195 | | | 1.20 ± 0.04 | 11–12 |
| | 5 | 210 | | | 1.27 ± 0.09 | 10–12 |
| Moran *et al* [20, 21] | 2 | 102 | Linear 35°C/min | Peak shape, based on Randall-Wilkins [18] | 1.47 | 18 |
| | 3 | 137 | | | 1.77 | 21 |
| | 5 | 183 | | | 2.20 | 23 |
| Miller and Bube [28] | 2 | 86 | Linear 13°C/min | Initial rise Garlick and Gibson [19] | 1.07 | — |
| | 5 | 177 | | | 1.59 | — |
| | 8 | 242 | | | 1.98 | — |
| Present study | 2 | 97 | Quadratic $1.3 \times 10^{-6}$ $(°C\ sec)^{-1}$ | First order Kelly-Laubitz [23] | 1.10 | 13 |
| | 3 | 137 | | | 1.21 | 13 |
| | 5 | 197 | | | 1.97 | 20 |
| | 8 | 265 | | | 1.96 | 17 |

1 to 8. Many of these peaks are useful for activation analysis, although peak 1 decays too rapidly at room temperature, and peaks 4 and 7 overlap peaks 5 and 8 too severely for accurate analysis.

The present results for LiF (TLD-100) are given in Table III in comparison with other values from the literature. Second order calculations (not shown) give very large values of both $E$ and $s$, and clearly do not apply to these peaks in LiF, in agreement with other work. In general, the values obtained in LiF in this work and those given in the literature agree less well with one another than in the previously discussed cases. This is true both with the peak temperatures and with the activation parameter values. Some of these discrepancies may be explained as due to poor thermal contact or other experimental reasons. Zimmerman *et al* [26] and Jackson and Harris [27] for example, used the isothermal decay method which has been considered to be less accurate than other methods due to the necessity of determining slopes. The values for the activation energies for peaks 2 and 8 in the present analysis do agree well with the initial rise values of Miller and Bube [28], but the other values obtained, and other results reported in the literature, generally show a wide variation for all peaks.

Moran *et al* [20, 21] recently used a phenomenological interpretation of TL glow-curve shapes to demonstrate how discrepancies in activation parameters determined from peak-shape analyses may be reconciled with more reasonable values. Such is the case with the present values of $E$ and $s$ for peak 5, where abnormally high values of $E$ and $s$ were consistently obtained. (Additional determinations of the activation energy for peak 5 using a linear heating rate and Chen's analysis gave results very close to those determined using the Kelly-Laubitz analysis.) Comparing the present results with the "initial rise" results of Miller and Bube [28], it is observed that peak 5 is about six degrees too narrow. That is, an arbitrary increase in the half width of this peak by about three degrees (keeping the same peak temperature) yields a result close to that of Miller and Bube. This half-width difference is much greater than the possible experimental error in the present work. This illustrates the basic problem of the peak-shape techniques, namely the inability to deal with small changes in line shape. This point has also been observed and discussed by Moran *et al* [20, 21].

These observations lead one to the conclusion that in addition to the theoretical questions which have been raised regarding activation parameters, serious experimental uncertainties are also involved. Peak shape methods are seen to be susceptible to errors due to changes in line shape. The "initial rise" method, on the other hand, has less to do with precise line shapes, and gives results which seem to be generally more consistent than those of other techniques. Due to theoretical uncertainties, however, it is doubtful whether even the initial rise method gives absolute values that have any real importance. For comparison purposes, it would probably be valid to compare values obtained by either method, provided that the samples and experimental conditions are equivalent. However, since values for comparable peaks seem to agree with one another to within 20% anyway, such comparisons may be of little use. It is apparent, therefore, that determinations of activation parameters in themselves are much less meaningful than observations of other TL characteristics.

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