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BETA DOSIMETRY USING PULSED LASER HEATING OF TLD MATERIALS*

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Abstract: Use of a pulsed CO_2 laser to heat the surface of hot-pressed LiF chips has been investigated. The thermoluminescent traps in the first 10 to 20 μm of depth may be read out with good efficiency, which will allow entrance dose and exit dose to be determined using a standard chip. These dose data can be used to calculate beta dose and gamma dose separately. Readout speed is estimated to be a few milliseconds per chip.

Introduction

Current personnel dosimetry used for betas in a mixed beta-gamma field relies upon TLD's with differential shielding. Various subtraction or ratioing techniques are then used to assess the beta exposure independently of the gamma-ray exposure. This methodology is basically similar to that historically used with film badges, but the film has been replaced by a TLD. The key to operation, of course, is the relatively large absorption coefficient shown by the betas with respect to the gamma-rays. This enhances the effects of differential shielding, but it also means that all detectors show significant gamma response, which limits the accuracy of any subtractive or ratiometric technique. It would be better if a means were available to determine the beta exposure as recorded by the detector (a TLD for example) independently of the gamma exposure. Various types of thin detectors that could be sandwiched together for exposure and separated for readout would be appropriate. A depth dose or absorption curve could be constructed from such an assembly which would then aid in separation of the more strongly attenuated beta dose deposition from that expected for gamma-caused deposition. Braunlich (Br81) has demonstrated such methods with micron thicknesses of specially prepared TLD material. It would appear that the TSEE phenomena could also be used for assessment of dose in a very thin layer of the detector material. Both these techniques will require

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new dosimeter materials however, in addition to some new apparatus for readout. Many organizations have resisted changeover to TLD's from film for financial reasons, and it seems unlikely that yet another new detector is the proper approach. This paper addresses the use of existing TLD materials, LiF chips in particular, as beta dosimeters by taking advantage of nonuniform heating with a pulsed laser to thermally dump only the near-surface traps.

Preliminary calculations as given below show that steep spatial gradients may be obtained in LiF chips with shaped pulses from readily available lasers. These steep gradients will allow only the thin entrance layer of TLD material to be heated sufficiently to be read out. Directing the laser at the exit face of the TLD could yield a measure of the more penetrating gamma exposure. The signal-to-noise ratio of this method is significantly better than the usual hot plate heating procedure, as pointed out by Braunlich (Br81). It is thus expected that adequate sensitivity can be obtained even for the thin sections heated via laser irradiation. Readout times of a few milliseconds are anticipated.

Use of a laser as a heat source represents a new addition to the equipment needed to read personnel badges. Perhaps a faster light integrator and some computer data collection methods will be required. However, the salient feature of this method is that it uses existing standard TLD's. These TLD's can still be read with the usual hot plate heaters for other applications. No new badges or special holders need be developed. And lastly, the speed of readout will be dramatically increased, which can lead to lower costs in handling of the dosimeters.

The following material is a theoretical investigation of the heating of hot-pressed LiF TLD chips with a pulsed CO_2 laser. A relatively simple model has been chosen, but it has proven adequate to demonstrate the general feasibility of this heating method. As will be seen, the temperature versus depth profiles are sharp enough that only a very thin surface layer is heated. Existing trap parameters (Fa78) and the calculated temperatures have been used to predict glow curves. These show that the principal LiF peak at $\sim$190°C can be evaluated with only minimal contributions from the other peaks. This system thus seems to be adequate for dosimetric purposes.

Temperature Response

The transient temperature response of a LiF chip irradiated by a laser pulse can be estimated by applying classic heat flow theory to a suitably simplified model. The model selected here is the opaque, passive, isotropic, semi-infinite solid (Ca59) which is reasonably faithful to the real system for brief infrared laser pulses and modest temperature rise. The solid is initially at uniform temperature throughout, and at zero time the opaque insulated surface, the plane $x = 0$, is exposed to the laser pulse. Isotherms are planes parallel to the surface, and the heat conduction equation reduces to the simple parabolic form with the solution given by (Da63)

$$U(x,t) = (\pi K \rho c)^{-\frac{1}{2}} \int_0^t H_a(t-\tau)\, \exp\left(-x^2/4K\tau\right)\, \tau^{-\frac{1}{2}}\, d\tau \qquad (1)$$

where

$U(x,t)$ = rise above initial temperature at time t and depth x

ρ and c = density and heat capacity, respectively, of the solid

K = thermal conductivity of the solid

$H_a(t)$ = irradiance (power per unit area) absorbed at the surface of the solid (H_a is always zero for $t < 0$)

κ = thermal diffusivity of the solid

Several pulse shapes, including rectangular, Gaussian, and triangular, have been analyzed in the course of our studies. Differences among these various shapes are not large, particularly at depths exceeding one relaxation length, ℓ. This latter parameter is defined through the full width of the pulse at half maximum (FWHM), η, as

$$\ell = (4\kappa\eta)^{\frac{1}{2}} \tag{2}$$

For present purposes, interesting features of laser heating of TLD's are illustrated using a triangular pulse of equal rise and fall times; this functional form is easily handled mathematically, while the temperature response is almost indistinguishable from the physically more realistic, but mathematically more awkward, Gaussian pulse. The absolute temperature of the chip, assumed initially at 293 K (20°C), is found from Eq. (1) to be:

$$T(x,t) = 293 + Q_a \cdot (\pi\eta K\rho c)^{-\frac{1}{2}} f(x,t) \tag{3}$$

where Q_a is the total energy absorbed per unit area of exposed TLD, the other parameters have been defined previously, and

$$f(x,t) = 0, \quad t \leq 0$$

$$= g(x,t), \quad 0 \leq t \leq t_p$$

$$= g(x,t) - 2g(x,t-t_p), \quad t_p \leq t \leq 2t_p$$

$$= g(x,t) - 2g(x,t-t_p) + g(x,t-2t_p), \quad t \geq 2t_p$$

where

t_p = time to peak of triangular pulse

$$g(x,t) = (4/3)\ (t/\eta)^{3/2} \left(6\sqrt{\pi}\ i^3\mathrm{erfc}\left(x/(4\kappa t)^{\frac{1}{2}}\right)\right)$$

Here, $i^3\mathrm{erfc}(z)$ is the third repeated integral of the complimentary error function (Ab65); the term in braces is exactly unity for $x = 0$ (i.e., the surface response). Solutions of Eq. (3) are shown in Figure 1 as absolute temperature versus time with depth as a parameter, and in Figure 2 as temperature versus depth with time as a parameter. These curves are specific for a pulse FWHM of 10^{-3} sec and a peak surface

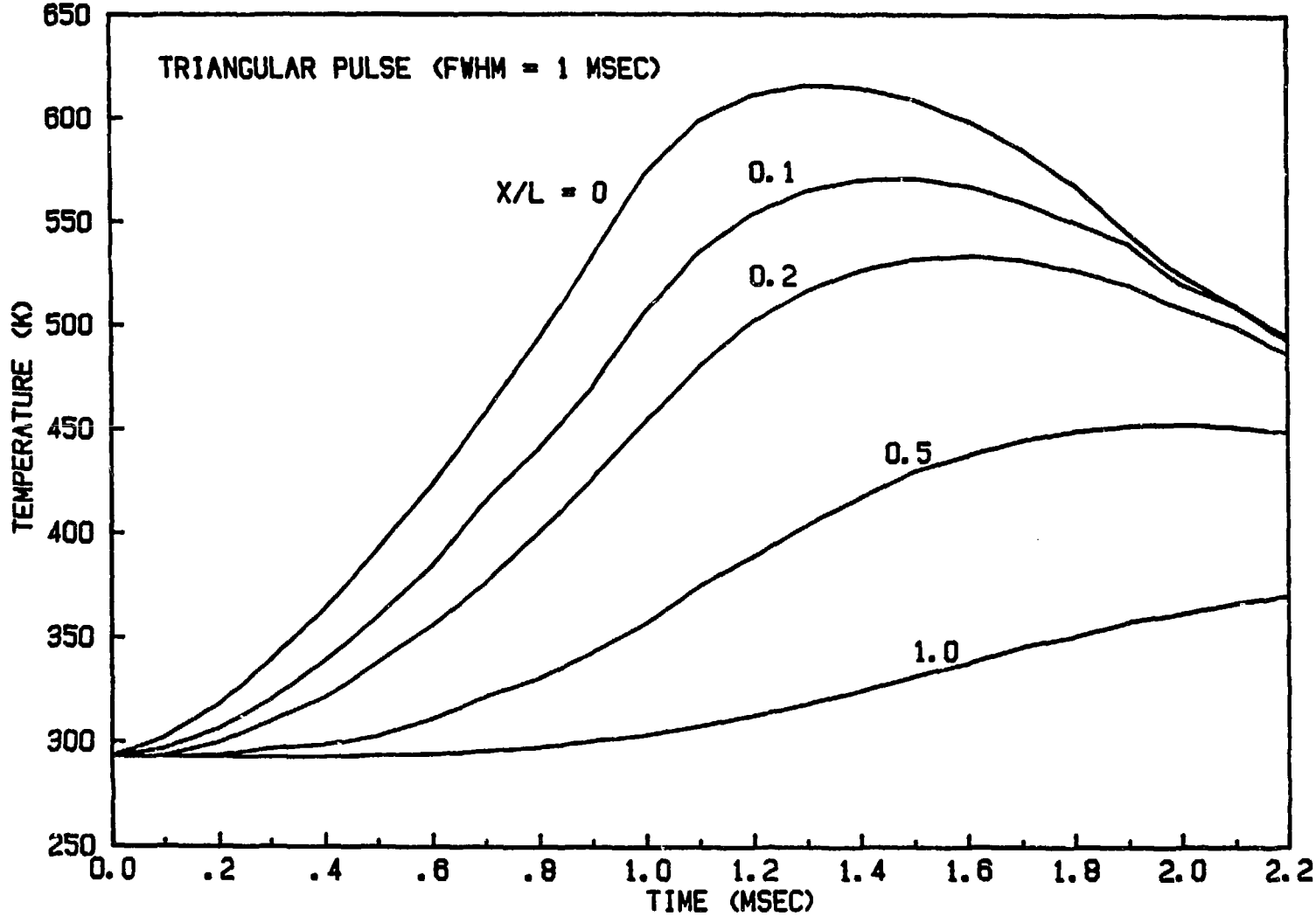

Figure 1. LiF temperature versus time, $\ell \simeq 170$ μm

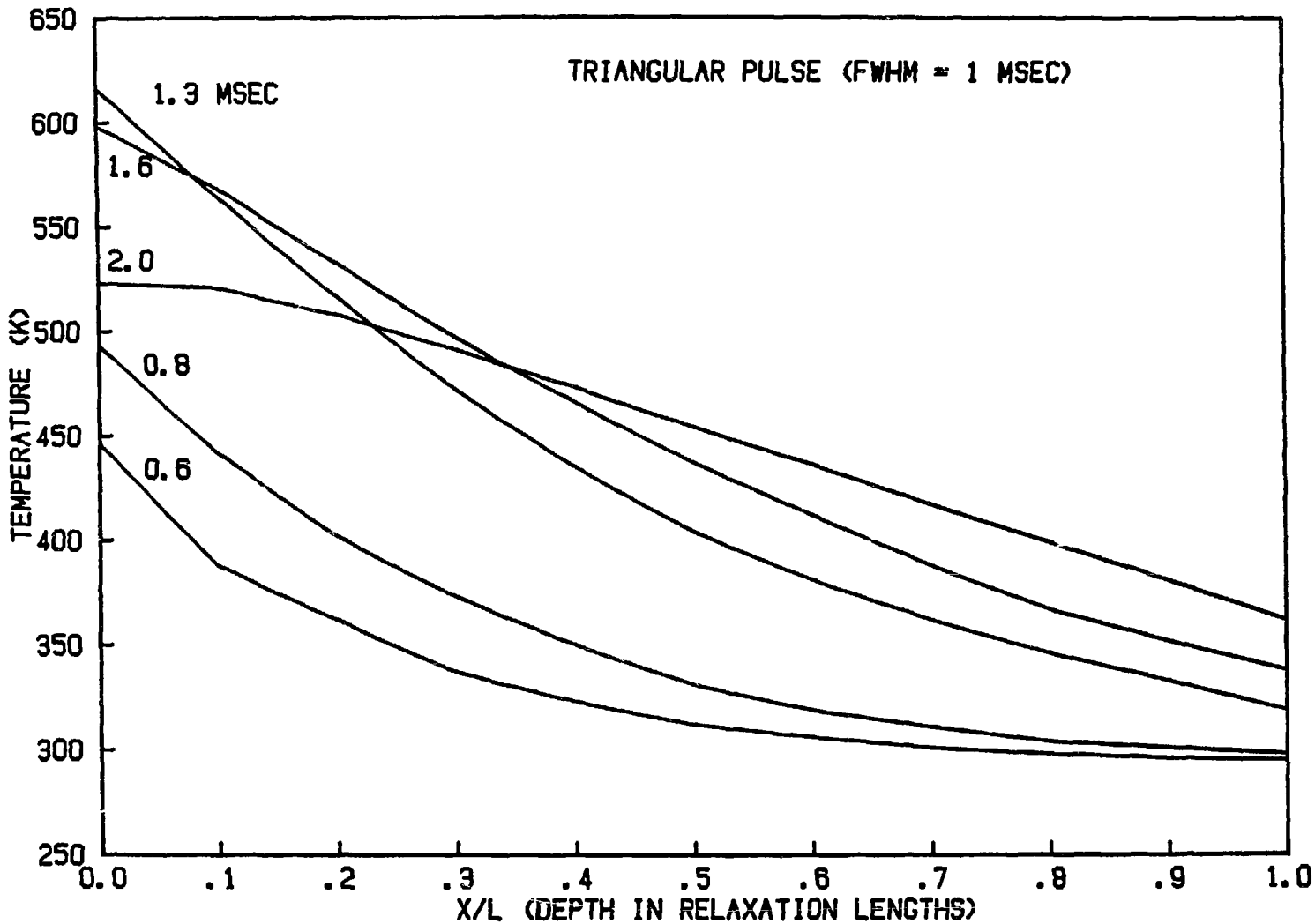

Figure 2. LiF temperature versus depth, $\ell \simeq 170$ μm

temperature of 616 K. The rationale for these particular choices will be explained in the following section.

At this point, we note that the depth parameter in Figure 1 and 2 is expressed in terms of the relaxation length, ℓ, which is defined by Eq. (2) as $(4\kappa\eta)^{\frac{1}{2}}$. The thermal diffusivity, κ, of LiF is 7 mm^2 sec^{-1}, (Br81) and, as just noted, for these curves, η is 10^{-3} sec. Hence, the relaxation length, ℓ, is approximately 170 μm. As will become apparent in the next section, essentially all of the thermoluminescent output is restricted to depths of the order of 10% of the relaxation length.

Thermoluminescence Kinetics

In analyzing the thermoluminescence of laser-heated TLD's, we follow the usual assumptions of first-order kinetics of each trap species,

$$I_i(x,t) = -B_i(dn/dt) = B_i\, n_i(x,t)\, \lambda_i(x,t) \tag{4}$$

where

I_i = TL output from a depth x at time t for the i^{th} trap species

n_i = trapped-charge concentration at x and t

$\lambda_i(x,t)$ = decay constant for the i^{th} trap at x and t

B = proportionality constant.

The rate constant is given by

$$\lambda_i(x,t) = s_i \exp\left(-E_i/kT(x,t)\right) \tag{5}$$

where s_i and E_j, the frequency factor and trap depth for the i^{th} trap are assumed independent of temperature and are single valued (Fa78); k is the Boltzman constant. Solving Eq. (4) for n, we can write

$$n_i(x,t) = n_{oi} \exp\left(-\int_o^t \lambda_i(x,v)\, dv\right) \tag{6}$$

where n_{oi} is the initial charge concentration in the i^{th} trap. The i^{th} glow curve can thus be written as

$$I_i(x,t) = \left(B_i n_{oi}\right) \lambda_i(x,t) \exp\left(-\int_o^t \lambda_i(x,v)\, dv\right) \tag{7}$$

where λ_i given by Eq. (5).

To select a reasonable set of parameters for this preliminary study, we first consider the trap parameters, s and E. Representative values for seven traps are shown in Table 1 (Fa78).

Table 1. Average values of trap parameters

Peak Number	Trap Depth E (eV)	Frequency Factor s (sec^{-1})
1	1.04	10^{14}
2	1.07	10^{13}
3	1.05	10^{11}
4	1.54	4×10^{15}
5	2.20	10^{22}
5a	1.61	10^{15}
6	1.70	10^{15}

Logarithms of rate constants (Eq. [5]) versus T^{-1} for each of these peaks are plotted in Figure 3. We note immediately that while the peak numbers are ordered sequentially by decreasing rate constant at the lower temperature associated with conventional thermoluminescent readout, this is not the case at the higher temperatures attainable with laser heating. Given that one can easily remove peaks 1 and 2 with a relatively low-temperature post-exposure anneal, then peak 5, the common "190°C dosimetric peak," becomes the most labile of those remaining. We can now use Figure 3 to estimate practical exposure parameters to read out this peak 5, relatively uncontaminated by other peaks, over appropriate depths into the TLD.

First, for best accuracy and precision, we ask that the dosimetric peak be essentially completely "glowed out." Referring to Eq. (6) this means that the integral of λ over the total temperature excursion should be at least 4 ($e^{-4} = 1.8 \times 10^{-2}$), which means that 98% of the associated traps have been emptied. Empirically, we have found for a variety of pulse shapes, amplitudes, and durations that when the product of the maximum rate constant (for maximum TLD temperature) and the laser pulse duration (FWHM) is about 10, then the integral of λ will indeed be $>\sim 4$.

As an application of this empirical finding, we note from Figure 3 that we can achieve a maximum rate constant for peak 5 of 10^7 sec^{-1} at $10^3/T = 1.35$, or a maximum temperature of about 740 K. To completely read out peak 5 at the surface, we now select a laser pulse width of 10^{-6} sec, to meet the criterion that $\lambda \cdot \eta \approx 10$.

Next, we note a second empirical observation. Again, for a variety of pulse parameters, the maximum rate constant at a depth of about 0.07 relaxation lengths into the chip is 10% of the surface maximum, and is 1% of the surface maximum at $\sim$0.14 relaxation lengths into the TLD. Recalling that the relaxation length is defined as $(4\kappa\eta)^{\frac{1}{2}}$, then for the 10^{-6} sec exposure,

$$\ell = \left((4)\ \left(7\ mm^2\ sec^{-1}\right)\ \left(10^{-6}\ sec\right)\right)^{\frac{1}{2}}$$

$$\approx 5\ \mu m$$

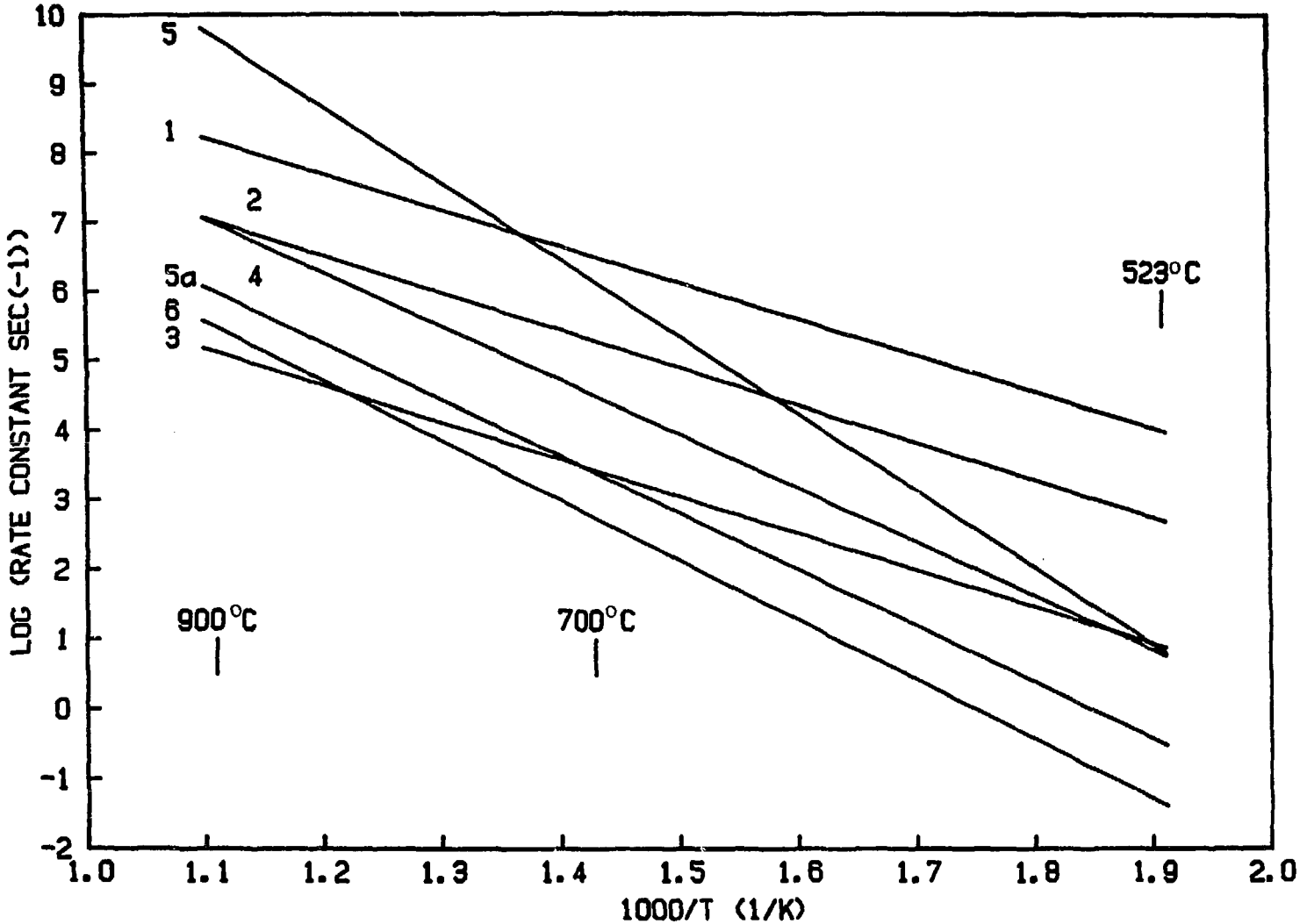

Figure 3. Rate constants for LiF (Fa78)

and the effective readout depth for this situation is only 5000Å, which is unreasonably shallow. While this consideration makes a 10^{-6} sec exposure unacceptable, we do note from Figure 3 that the maximum rate constant for the nearest competing glow peak, No. 4, is almost two orders of magnitude below that for peak 5, so we have achieved a relatively "clean" single-peak readout.

We now approach this problem of selecting reasonable exposure parameters from the opposite direction. We first select a readout depth of the order of 10 μm, which is a reasonable fraction of the extrapolated range of operationally interesting beta particles in LiF. Since this will be about 10% of the relaxation length, it follows from Eq. (2) that the pulse duration (FWHM) should be of the order of 10^{-3} sec. For essentially complete surface readout of peak 5, the maximum rate constant should then be 10^4 sec^{-1}. From Figure 3, this calls for a peak surface temperature of about 616 K, or a 323 K rise above a 20°C initial temperature. At this point, the maximum rate constant of peak 4 is only one order of magnitude below that for peak 5, so one must accept some contribution to the final glow curve from peak 4. Emission from peaks 3, 5a, and 6 will not be measurable.

Glow Curves

Suggested exposure parameters established to this point are:

$$\text{pulse width (FWHM)} = 10^{-3} \text{ sec}$$

$$\text{maximum surface temperature} = 616 \text{ K}$$

Solution of Eq. (3) gives temperatures to calculate glow curves by Eq. (7). Relative glow curves for peak 5 at the TLD surface and at depths of 12 μm and 24 μm are shown in Figure 4, together with the peak 4 glow curve from the surface. The "sum peak" shown in Figure 4 is only meant to suggest the general form of the total glow curve; a more careful analysis would involve an integration over depth rather than this simple four-component summation. We have also assumed here that the initial trapped-charge concentrations and conversion efficiencies of peaks 4 and 5 are the same.

The essential point of this simple analysis is that the composite glow curve of a 10^{-3} sec pulsed-laser-heated TLD is a reasonably clean representation of the response of a single thermoluminescent peak – peak 5 – over roughly 10 μm depth into the TLD chip. For the 616 K maximum surface temperature envisioned in this analysis, peak 5 traps will be substantially emptied completely in this surface layer, yielding dosimetric accuracy and precision as well as calibration stability.

In passing, we note that for a LiF density of 2.4 g cm^{-3} and heat capacity of 0.2 cal g^{-1} K^{-1}, (Br81) the 616 K maximum surface temperature in a 10^{-3} sec FWHM pulse requires peak laser irradiance of about 7 kW/cm^2 on the chip, or an energy density of about 7 J/cm^2. These are high but achievable performance figures.

Summary and Conclusions

This study has shown that surface heating of typical hot-pressed chips of LiF can be achieved with a CO_2 laser. A triangular shaped laser pulse of FWHM of 1 msec

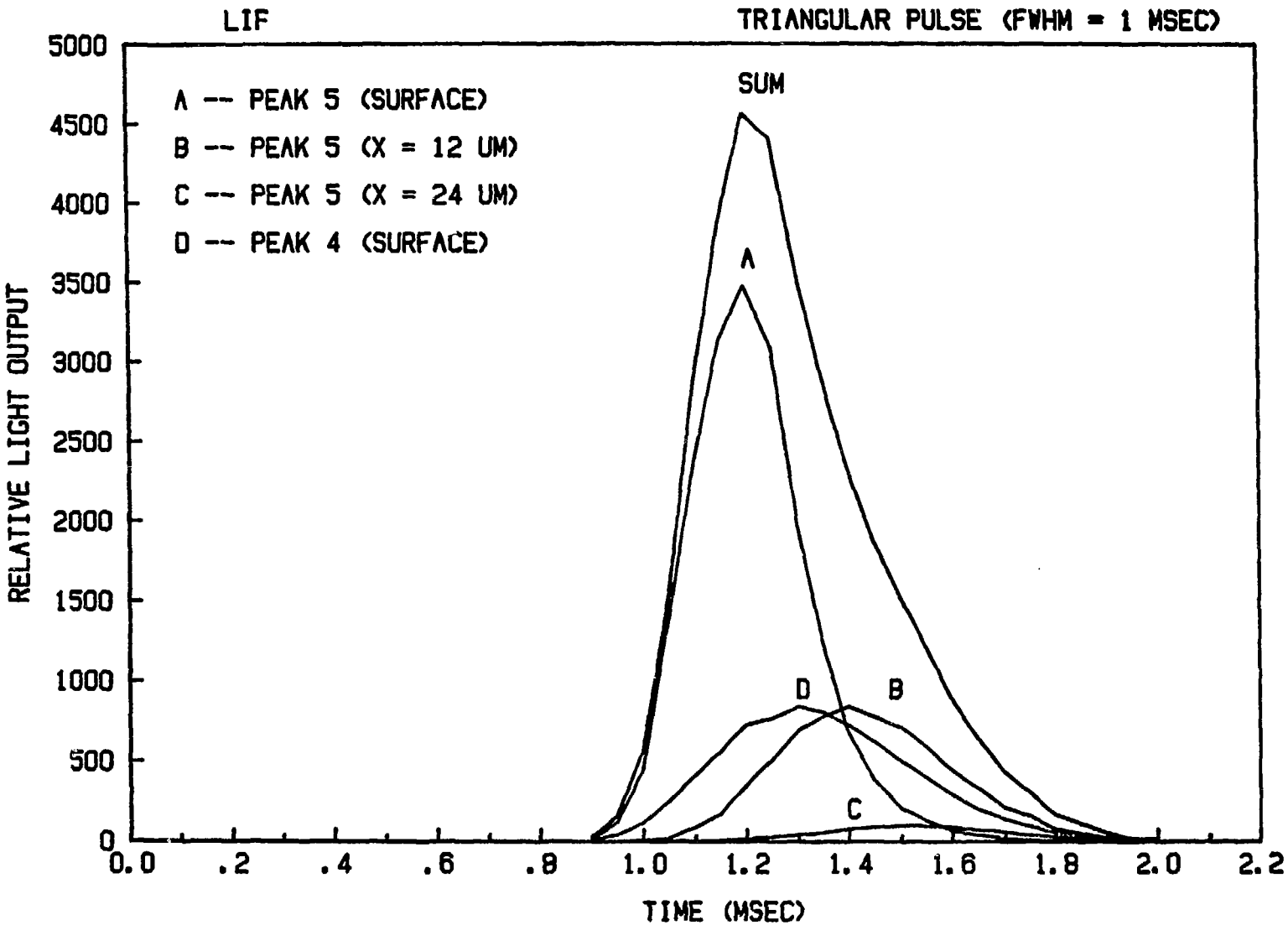

Figure 4. LiF glow curves expected for pulsed laser heating

capable of delivering 7 kW/cm^2 to the chip surface will result in temperatures of 616 K at this surface. The temperature gradients caused are steep and will allow only $\approx$20 μm of material to be heated sufficiently to dump traps of dosimetric interest (peak 5, 190°C) at any significant rate.

This heating procedure will result in estimation of the dose in a very thin layer of LiF. If the usual hot-pressed LiF chip is exposed to a mixed beta-gamma field, a thermoluminescent gradient will be produced within the chip. The pulsed laser heatir technique will allow both entrance and exit thermoluminescence to be determined. These data can be used to estimate beta and gamma dose separately.

A secondary benefit of the laser heating technique is the speed with which a typical TLD is evaluated. Since heating is accomplished within a few milliseconds, it seems feasible to suggest reading both sides of every TLD. This will result in two data points per TLD and could be used to flag beta exposures or enhance gamma precision.

Note that these calculations have been based upon an "opaque, passive, isotopic, semi-infinite solid." Of these conditions only the first is violated to any significant extent. Calculations incorporating a more precise description of the absorption characteristics of LiF for the 10.6 μ laser pulse are underway. The variation with depth of the beta-induced thermoluminescence will also be modeled.

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