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ABSTRACT

In ~~This paper presents~~ an experimental study on PTTL in LiF TLD-100 sample, PTTL is studied as a function of residual TL, wave length of UV light and temperature of sample during UV exposure. FOLLOWING results are obtained

1) The relative intensities of individual glow peaks in PTTL glow curve are remarkably different from those of X or gamma ray induced TL glow curve. The intensity changes are so drastic that peaks 1, 2 and 3 can be stronger than peak 5

2) The efficiency of transfer per unit photon fluence has a maximum value for 230 - 240nm light in all the sample studied, which indicates that one of the residual traps from where charge transfer occurs resembles closely F and Z_3 centres. Charge transfer from another residual trap is most effective with 290nm light

3) The intensity of PTTL is not proportional to the total residual TL of a sample. It appears that the efficiency of transfer from different types of residual traps is different. Also different residual traps redistribute their optically released charge carriers preferentially amongst the available empty traps.

4) The transfer efficiency is very much dependent on temperature of the sample during UV exposure. This shows that charge release from residual trap is partly by UV photon energy & partly under thermal stimulation. The UV photon excites the charge carrier from residual trap to an intermediate level close to but below the free state, from where thermal stimulation liberates it into the free state. This thermal energy gap is computed to be 0.12 eV.

1 INTRODUCTION

The stimulation of thermoluminescence (TL) by ultraviolet (UV) light in samples partially bleached (thermally) after a high gamma ray exposure has been observed to be a common phenomenon in most TL phosphors[1-7]. Sunta[1] further showed in natural CaF_2 that TL intensity can be transferred from a given glow peak to other glow peaks appearing at lower as well as higher temperatures. The study of this phenomenon can contribute important information for understanding the mechanism of TL as well as for application in radiation dosimetry. UV induced TL in LiF TLD 100 was first studied by Pearson and Cameron[6,7]. Later Podgorsak et al[8] and Sunta et al[9] have extended the UV induced TL glow curve to liquid nitrogen temperature at lower end and up to 400°C on higher temperature side respectively. Although it is generally known that soft-UV induced TL is due to transfer of charge carriers from deep residual traps, it is not known what the deep traps actually are and how does the transfer take place. In the present paper a detailed experimental study is carried out on UV induced TL in LiF TLD 100 and an attempt is made to explain the transfer process in the light of the observations.

2 *Experimental Procedure*

A powder sample of LiF TLD 100 as received from Harshaw Chemical Co. was annealed at 400°C for one hour and exposed to about 10^5 R of ^{60}Co gamma rays. It was then divided into three parts and annealed at three different temperatures in order to have different amounts of residual TL in the samples. Part 1 was annealed at 228°C to bleach out peaks populary numbered as 1 to 5. Part 2 was annealed at 322° C. Part 3 was annealed at 360° C to ensure that only one glow peak i.e. that of highest temperature[9,10] remains residual in the sample. Each of these annealings were done for one hour in an air oven. Residual TL of each of these batches was recorded by using Harshaw 2000 TLD reader whose pan heater supply was replaced by a linear programmer capable of heating up to 550° C. UV induced TL glow curves were read for the three sample batches as a function of λ of incident light as well as temperature during UV exposure. All UV exposures were carried out using monochromatic lights of band width 2.4nm (full width at half maximum). The light source used was a Xe lamp (Bausch & Lomb. Cat. N° 33 86 20 01) with a high efficiency UV monochromator (B & L cat. N° 33 86-25 01). Light intensities were measured using an EG & G Radiometer model 580 placed at a distance behind the sample position and corrected for the difference in position. The observed peak heights in the glow curve were normalised to obtain the TL intensity per unit photon fluence at each wave length.

The study of temperature depedence was carried out with the same sample of TLD 100 except that it was ground very finely with a mortor and pestle. The annealing treatments and gamma exposures were same as described above. In this experiment 5 mg of fine sample powder was deposited on the reader pan by sedimentation in alcohol and drying it at about 100°C. This made a thin uniform layer in close contact with the pan, so that the sample could be assumed to have the same temperature as that of the pan, which was measured by a spot-welded thermocouple. UV exposures were given with 230 nm monochromatic light for 15 seconds at various steady temperatures of the pan from 25°C to 200°C and TL was measured after exposure at each temperature.

3. Results and Discussions

3.1. Effects of Residual TL on UV induced glow curve shapes and intensities

Fig. 1 shows the residual TL glow curves of the three samples namely those annealed at 228, 322 and 360° C after 10^5 R gamma ray exposure. The relative integrated light intensities under these curves are in the ratio of 50.6:3.7:1. As expected the residual TL intensity falls sharply as the temperature of post gamma exposure annealing is increased. Fig. 2a shows three typical glow curves after 230 nm UV exposure of the samples. For the purpose of comparison a glow curve produced by ^{60}Co gamma exposure also is given. This figure demonstrates very significant changes in the structure of the glow curves produced by UV exposure in comparison to that of gamma exposed samples. Firstly peak n° 4 is absent. This is in agreement with Mayhugh et al[11] and Sunta et al[9]. Secondly relative intensities in individual glow peaks changes remarkably with change in temperature of post gamma exposure annealing. These changes are specially more in 228°C annealed sample where peak N° 5 appears with weakest intensity in comparison to other peaks. Fig. 2b shows the glow curves with 290 nm exposure, where again 228°C annealed sample shows a complete reversal in the intensity distribution. Drastic changes in glow curve shapes of LiF have been observed earlier by Zimmerman et al[12] due to the effect of pre exposure annnealing temperatures. In the present case the observed changes are related to residual TL.

Since TL can be induced in LiF by soft UV only if the sample has been given a pre gamma exposure and then annealed at a temperature less than 400°C[9,10] keeping some high temperature traps still filled, it is reasonable to assume that such TL is indeed due to transfer of charge carriers from high temperature stable traps to those related with the usually observed 5 glow peaks The present results show that the transfer process is not of simple type in which optically liberated charge carriers would be recaptured in the empty traps in the same proportion as during X or gamma ray exposure It appears that the transfer from a residually filled trap to empty traps is of preferential nature i.e a charge carrier freed from a given trap may be selectively captured in particular category of traps rather than being equally attracted to all the avaible empty traps For example in case of transfer form residual TL peaks appearing above 400°C (Fig 1 curves B and C) the recapture probability is maximun for traps of peak 5 and decreases in order of peaks 3, 2 and 1 This creates an intensity distribution in UV induced glow curve nearly similar to that of gamma induced glow curve. (Fig 2a and b, curves 2 and 3) But on the other hand when the RTL extends to 300°C or less (228°C annealed sample) the transfer is more to peaks 1, 2, 3 (in this order) and least to peak 5 (Fig 2, a & b curve 4). Such preferential transfer would be possible if the residual parent trap and the capturing trap are physically close to each other

The overall intensity of the glow curve produced by same incident 230 nm UV photon fluence in 228°C annealed sample is less than half of that in 322°C annealed sample, although the total residual TL in the former is as much as 13 times higher than in the latter This shows that the presence of extra residual TL in 228°C annealed sample (apart from that of above 322°C, which is common in both) is unfavourable for transfer of charge carriers from residual traps stable above 322°C to those corresponding to usual five glow peaks In particular transfer to peak N° 5 is least favoured

3.2 TL Intensity as a Function of Incident UV wave-length

In Fig 3a,b and c are plotted the TL peak intensities of individual glow peaks as a function of λ incident UV light for the three samples Each of the curves has a maximum at 230 240nm region Curves of Fig 3a which correspond to the sample annealed at 228°C have in addition another hump in the region of 290nm The maxima in the curves should mean that the efficiency of transfer at corresponding wave lengths is maximum The most prominent peak at 230-240nm lies between the F and Z_3 absorption peaks of LiF These absorption bands appear with significant intensity in gamma irradiated LiF crystals annealed between 200 and 400°C[13]. This indicates that the residual trap from where charge carriers might be getting transferred may be either F and Z_3 centres or another centre closely related with them The additional indication of a hump at 290nm in Fig 3a, would mean that in case of 228°C annealed sample, the residual traps of stability between 228 and 322°C transfer their charge most efficiently under exposure to 290nm UV The glow peak structure for this case (Fig. 2b curve 4) further suggests that transfer is preferential to peaks 1, 2, 3 and 5 in order of decreasing preference. However no corresponding absortion band has been observed in 290nm region. There is over tenfold higher overall TL intensity induced by 290nm UV in 228°C annealed sample than in 322 and 360°C annealed ones It means that with this wave length the transfer is mainly from traps stable between 228 and 322°C, where as by 230 240nm UV the transfer takes place principally from such traps which are stable above 322°C The relative integrated residual TL intensities as well as the UV induced TL peak heights are tabulated in table 1, which bear out the preferential nature of transfer phenomenon.

3.3. Temperature Dependence of UV Induced TL

When UV exposures of the samples were carried out at different temperatures, it was observed that the induced TL intensity increases with temperature of the sample until the TL peak begins to bleach out thermally. Fig 4 shows the results. This observation shows that temperature dependence should be taken care of if TL phosphor is to be used for UV dosimetry. It was observed that intensity of UV induced peak 5 can be as much as 5 times stronger at 150°C sample temperature than at room temperature for the same UV exposure. This kind of behaviour has already been reported earlier in natural CaF_2[14]. Increased stimulation of TL with increasing temperature would mean that release of charge carriers from the parent residual trap is partly due to thermal activation in addition to optical activation under incident UV photons. If this is actually so, the transfer efficiency from the residual trap and consequently the observed transferred TL intensity should increase with temperature according to Boltzmann's relation for population in thermally excited states i.e.

$$I \alpha \exp [-\Delta E/kT]$$

where I should be TL intensity, ΔE thermal energy gap from optically excited state, to free state, k Boltzmann's constant and T absolute temperature. The total activation energy to make a charge carrier free from a deep residual parent trap before getting retrapped in other vacant traps would be equal to energy of UV photon plus ΔE. The above relation would give a straight line with negative slope $\Delta E/k$ if ln I is plotted against $\frac{1}{T}$. One can determine the value of ΔE from such a graph. Actual plotting shows that this relation does hold good with $\Delta E = 0.12$eV for the sample annealed at 360°C (Fig 5). However for the other two samples we do not get a straight line. It is because more thanone kind of traps are involved in those samples, whereas in 360°C annealed sample only one kind of traps, which are responsible for TL peak of highest temperature are residual so that only one activation energy would be involved.

Table I

Relative TL peak heights in UV induced glow curves of the three samples

<table>
<tr><td colspan="2">Annealing temps (°C) of the samples</td><td>228</td><td>322</td><td>360</td></tr>
<tr><td colspan="2">Relative Integrated Intensity of residual TL</td><td>50.7</td><td>3.7</td><td>1</td></tr>
<tr><td rowspan="4">Relative peak heights normalised to unit photon fluence incident on the sample</td><td>Peak 5 height with 235 nm (10^{-19} Amps)</td><td>0.84
(0.51)</td><td>2.6
(2.1)</td><td>1.25
(1)</td></tr>
<tr><td>Peak 2 height with 235 nm (10^{-19} Amps)</td><td>1.2
3.16</td><td>1
(2.6)</td><td>0.38
(1)</td></tr>
<tr><td>Peak 5 height with 290 nm (10^{-22} Amps)</td><td>53
(21.2)</td><td>10
(4)</td><td>2.5
(1)</td></tr>
<tr><td>Peak 2 height with 290 nm (10^{-27} Amps)</td><td>140</td><td>5</td><td>1</td></tr>
</table>

Note: Values within brackets are normalised with respect to sample with least residual TL (i.e.360°C annealed)

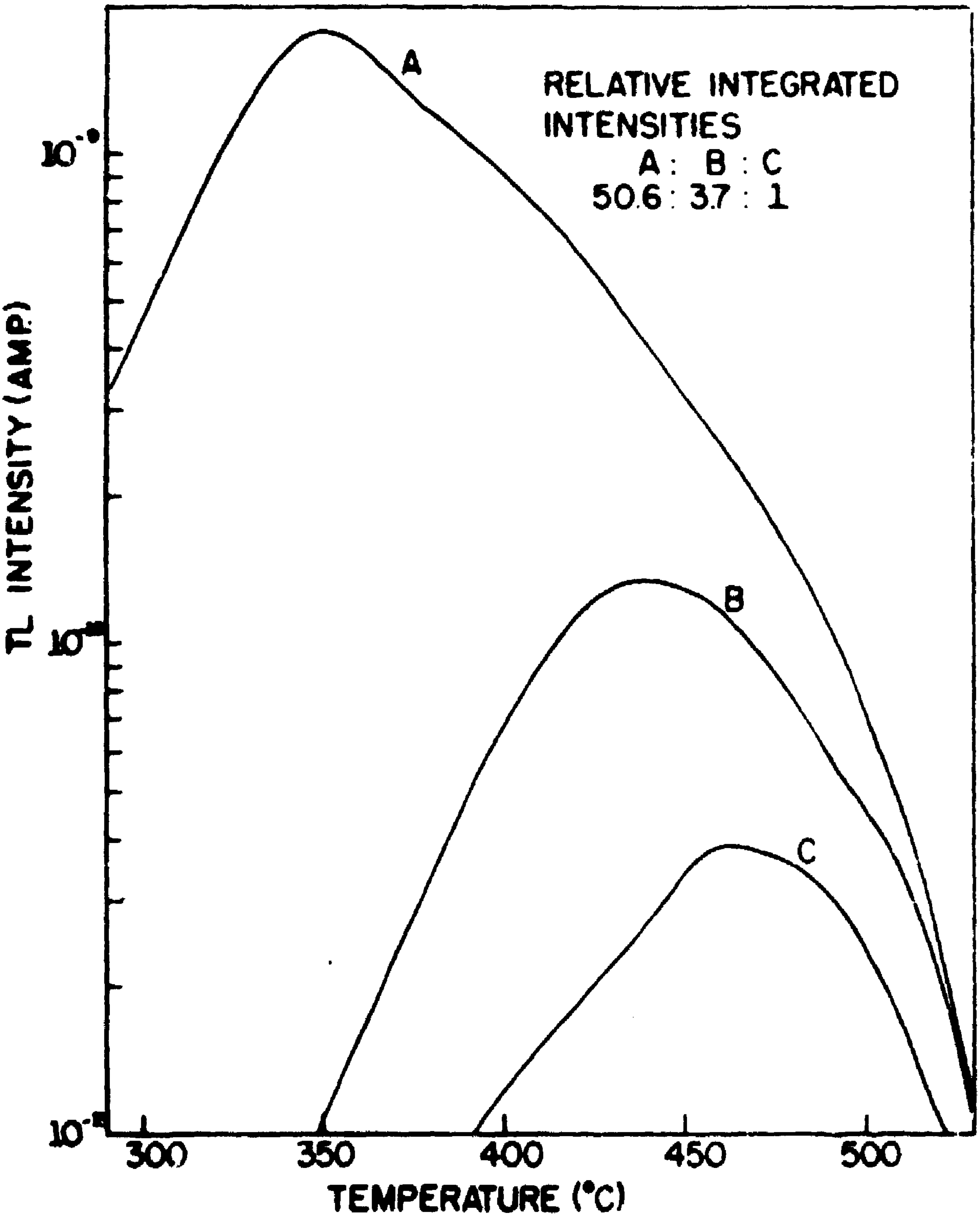

Fig.1 Residual TL glow curves of LiFTLD-100 exposed to 10^{6} R and annealed at A, 228°, B, 322° and C, 360°C respectively.

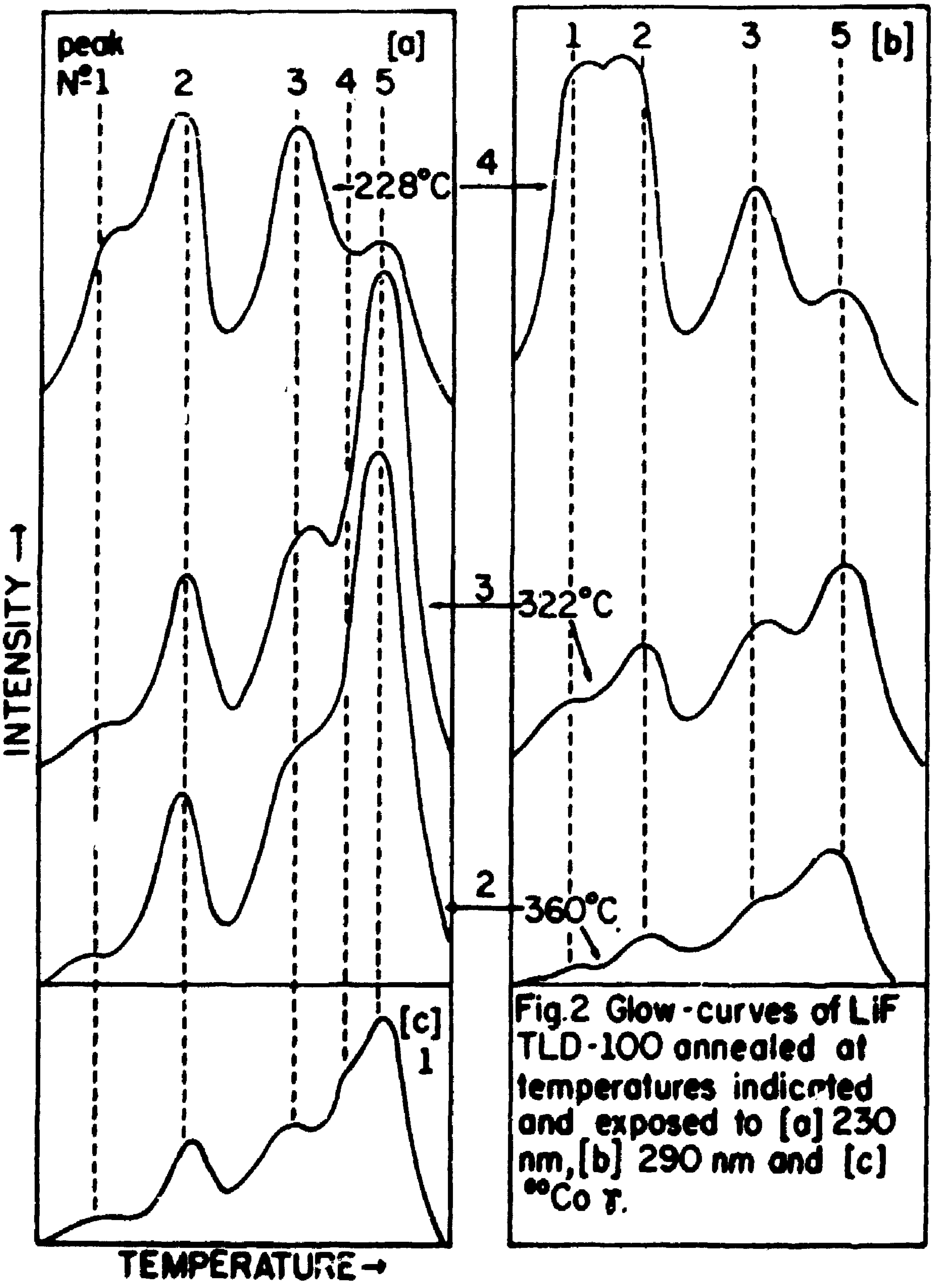

Fig.2 Glow-curves of LiF TLD-100 annealed at temperatures indicated and exposed to [a] 230 nm, [b] 290 nm and [c] ^{60}Co γ.

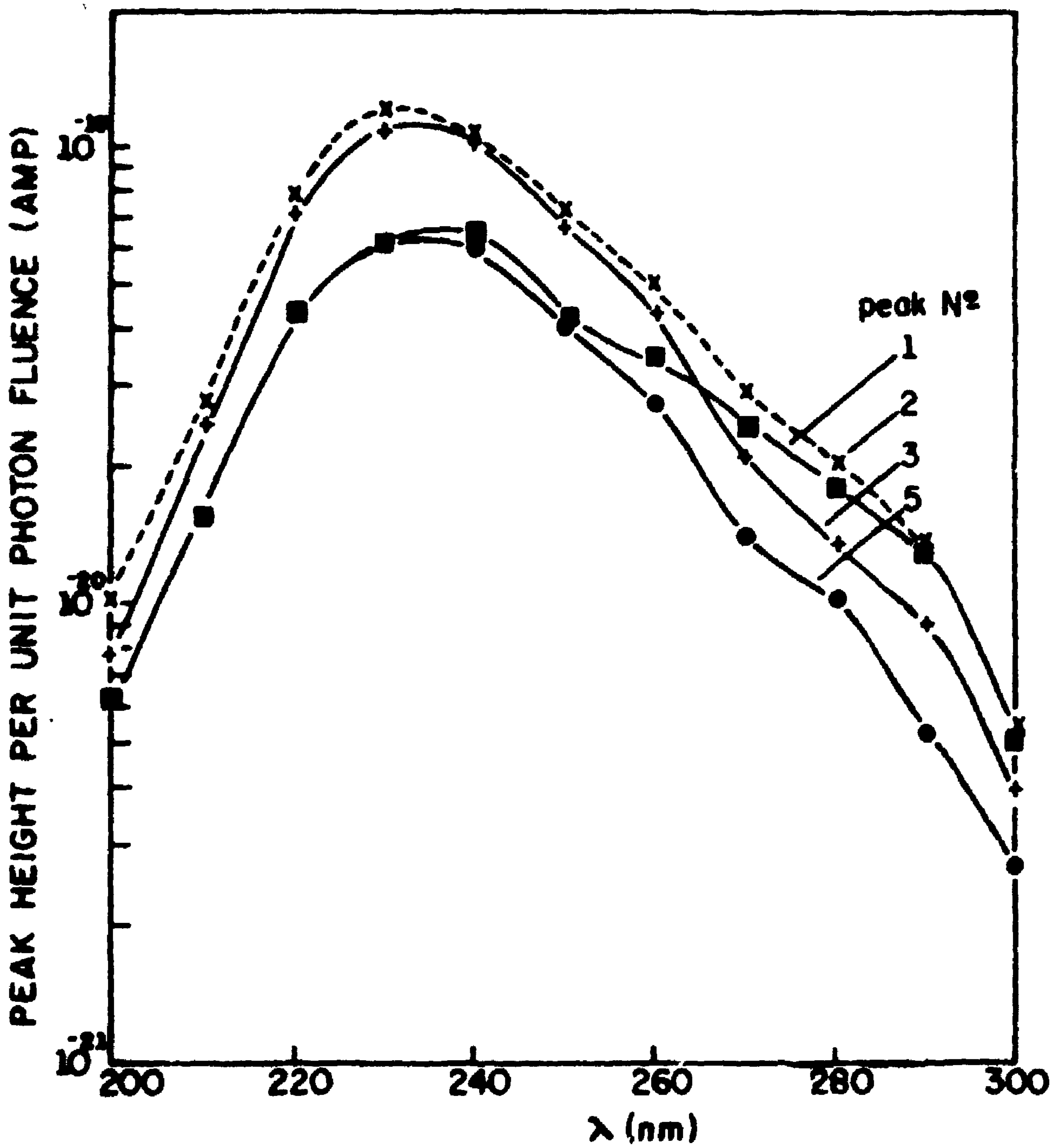

Fig. 3A Wave-length dependence of intensity of UV induced TL peaks in 228°C annealed sample.

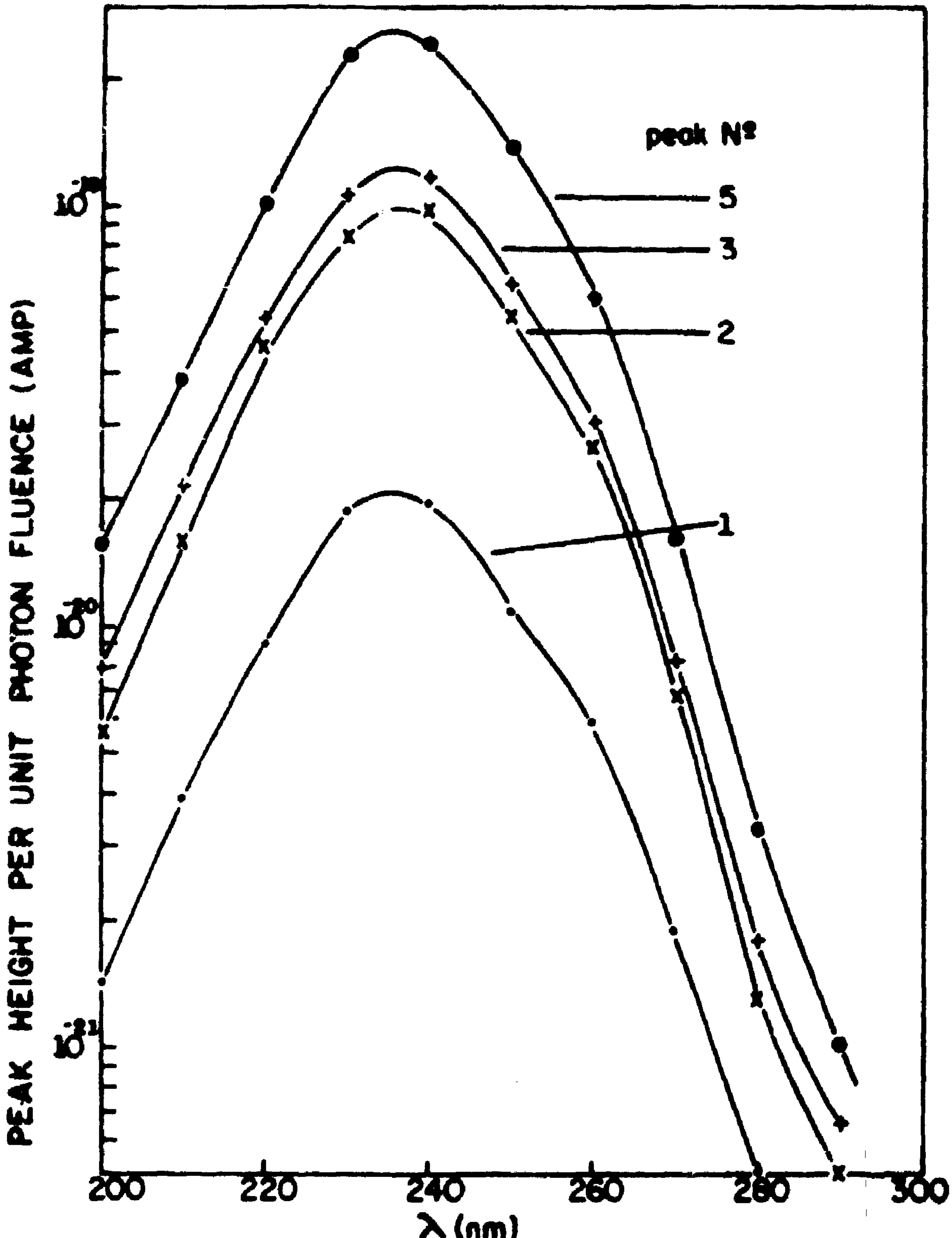

Fig.3B. λ Dependence of UV induced TL of 322°C annealed sample.

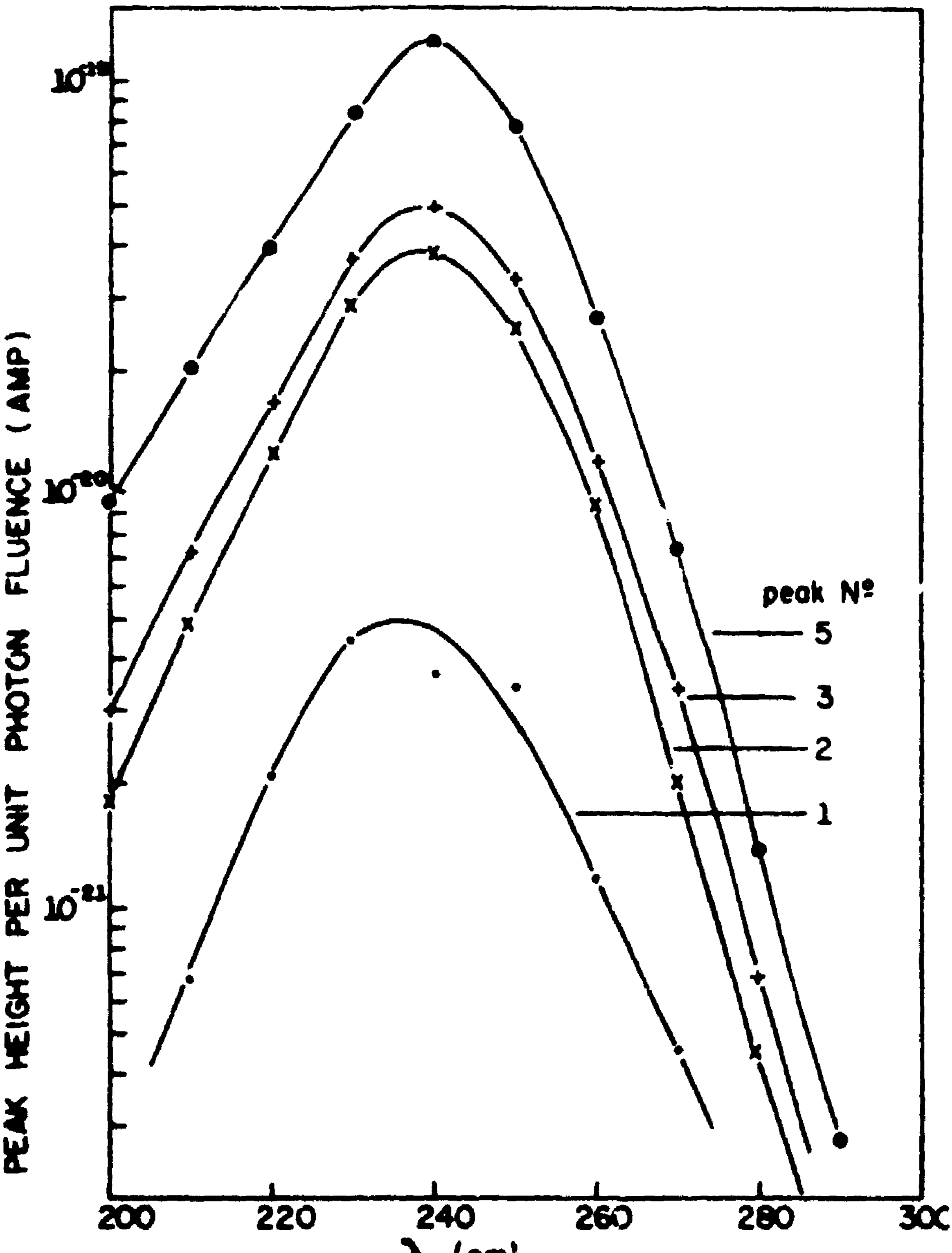

Fig. 3C. λ Dependence of UV induced TL in 360°C annealed sample.

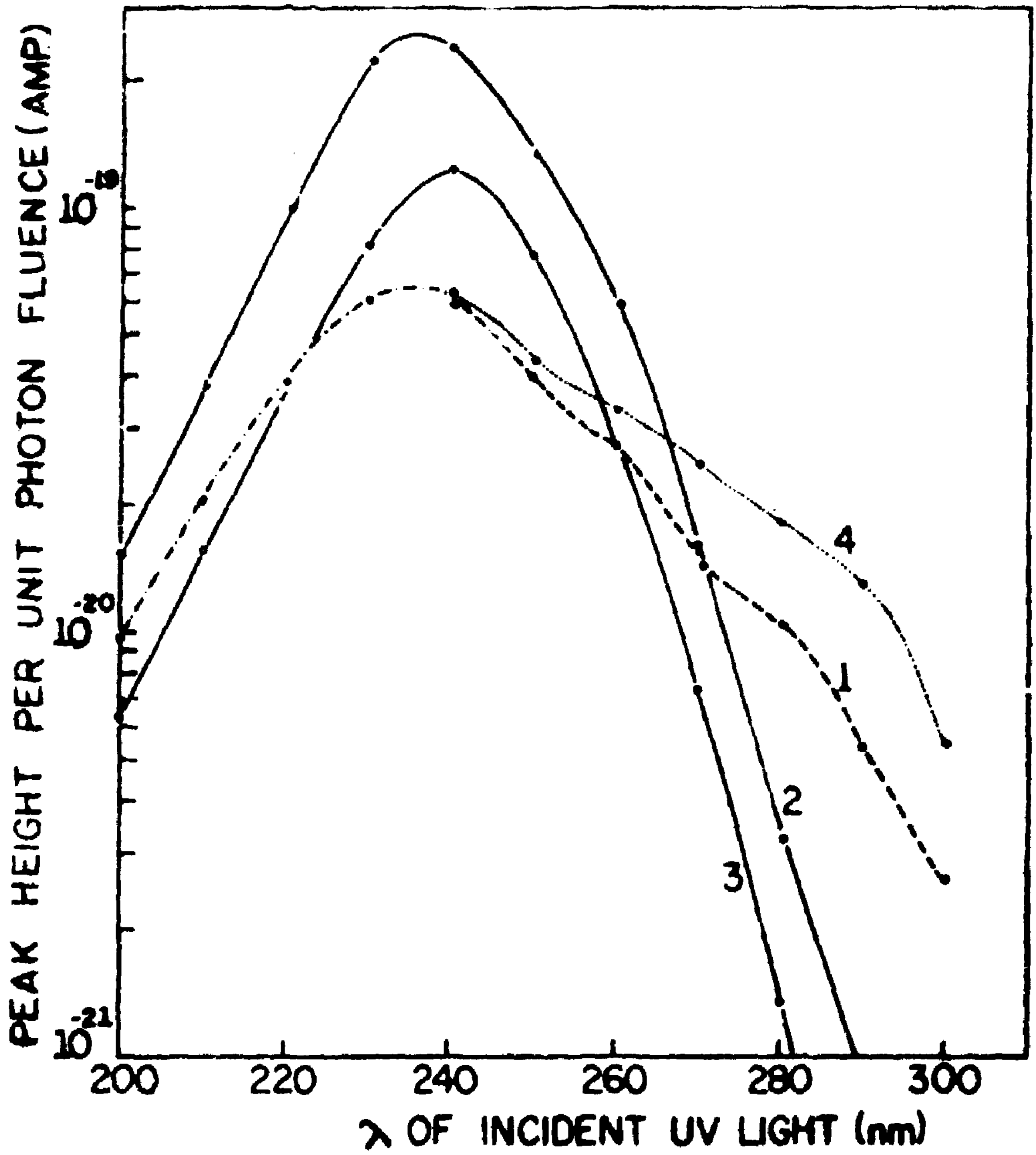

Fig.30. Wave-length dependence of UV induced TL peak 5 of LiF TLD-100 samples heated at 1) 228°C 2) 322°C, 3) 360°C, 4) TL peak1 of sample heated at 228°C.

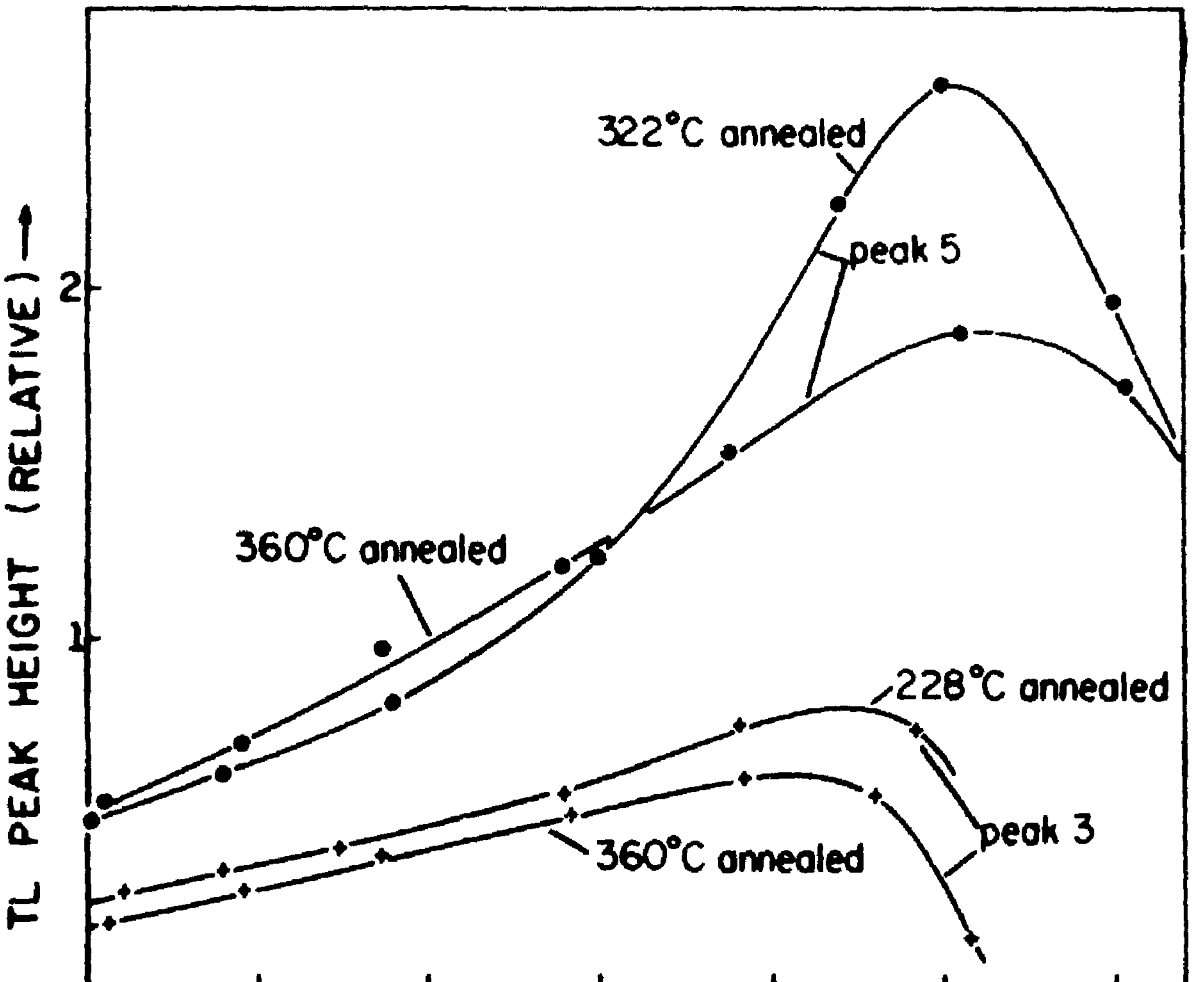

Fig.4 Temperature dependence of 230nm UV induced TL intensity. Peak 5 was not resolved in 228°C annealed sample.

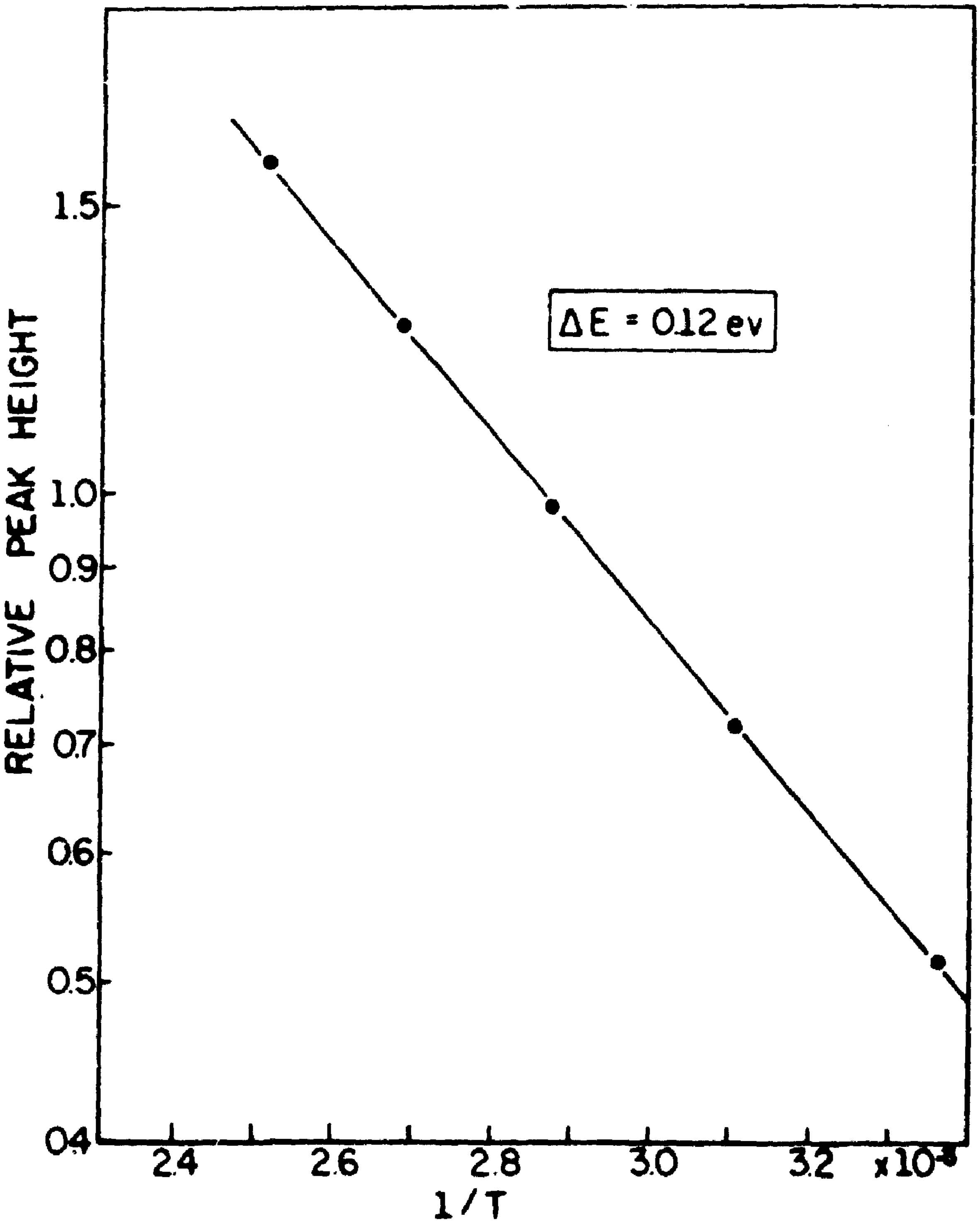

Fig.5 I vs. 1/T for peak 5 of 360°C annealed sample Exposure by 230nm light.

RESUMO

Este trabalho apresenta um estudo experimental sobre PTTL em LiF (TLD 100). PTTL foi estudada em função de TL residual, comprimento de onda da luz UV e temperatura da amostra durante a exposição UV. Foram obtidos os seguintes resultados:

1) As intensidades relativas dos picos individuais na curva de emissão PTTL são acentuadamente diferentes do caso de curva de emissão TL induzida por raios X ou gamma. As variações da intensidade são tão drásticas que picos 1, 2 e 3 podem ser maiores que o pico 5.

2) A eficiência de transferência por unidade de fluência de foton tem um valor maximo para a luz de 230-240 nm, em todas as amostras estudadas, o que indica que uma das armadilhas residuais da qual se da a transferência de carga, tem uma semelhança muito grande com centros F e Z_3. A transferência de carga de outra armadilha residual e mais efetiva para a luz de 290 nm.

3) A intensidade de PTTL não é proporcional a TL residual total de uma amostra. É aparente que a eficiência e transferencia dos diferentes tipos de armadilhas redistribuem os transportadores de carga oticamente liberados entre as armadilhas vazias disponiveis, desigualmente.

RÉSUMÉ

Ce travail presente une etude expérimental sur PTTL em LiF (TLD 100). PTTL est etudiee en fonction de la TL résiduelle, du longeur d'onde de la lumière ultraviolette et de la temperature de l'échantillon pendant l'exposition à l UV. Les suivant resultats sont obtenus:

1) Les intensités relatives des pics individuel de la curve d'emission PTTL sont remarquablement differentes de celles de la curve d'emission TL induite par les rayonements X ou gamma. Les changements d'intensite sont tellement drastique, que les pics 1, 2 et 3 peuvent être plus grand que le pic 5.

2) L'efficacité de transffèrement par unité de la fluence du photon possede un maximum pour la lumiere de 230-240 nm pour tous les échantillons etudiés; c'est une indication que l une des pieges residuelles, de laquelle le transfert de la charge a lieu, resemble a centres F ou Z_3. Le Transfert de la charge d'autre piège résiduelle est plus efficace avec la lumiere de 290 nm.

3) L intensite de PTTL n'est pas proportionelle a la TL residuelle totale d un echantillon. Apparemment, l efficacite du transfert de different type des pieges residuelles est differente. Aussi, les pieges residuelles differentes redistribuent les porteurs de la charge liberees optiquement non egalement entre les pieges vides disponibles.

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