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Theoretical Studies on the Physical and Environmental Factors Which Govern the Thermal Fading of Thermoluminescence Signal.

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ABSTRACT

Thermoluminescence solid-state detector is widely used to determine the dose in personnel and environmental monitoring for radiation protection purposes, for instance in the field of nuclear power production, medicine and research. However, thermal fading is a limiting factor for a long-term application, especially where temperature is changing significantly during the accumulation period. This paper studied the influence of temperature and duration of storage after irradiation on the thermal fading of the TL signal. Also, this paper discussed the dependence of the thermal fading on the trap parameters of TL glow peak. The most important parameters, which were considered here include the order of kinetics b, the depth of the trap level E (eV) and the frequency factor S (s-1). The dependence of the thermal fading on thermal stability parameters, namely trap depths and frequency factors for the glow peaks is discussed.

The variation of the thermal fading as a function of the order of kinetics is demonstrated. In addition, this paper discussed the dependence of the thermal fading on the absorbed dose in case of first-, second- and general-order kinetics. The above-mentioned studies were arranged considering the models of first-, second- and general-order of kinetics.

INTRODUCTION

Many radiation detectors have been developed over the last few decades and some are being used routinely for personnel dose control. One of them is the thermoluminescence (TL) solid state detector. This detector emits light when heated after exposure to radiation. It is widely used to determine the dose in personnel and environmental monitoring for radiationprotection purposes, for instance in the field of nuclear power production, medicine and research [1, 2, 3]. However, fading is a limiting factor for along term application, especially where temperature is changing significantly during the accumulation period. The thermal fading of stored signal occurs due to loss of the electrons from traps and the redistribution of trap due to migration and aggregation [3]. Hence, temperature is normally responsible for the fading, but other factors such as time of storage can also influence the loss of latent information in the TL material.

An important consideration in the choice of a TLD detector is how stable the signal is in the medium in which dosimeter operated. Studies on the thermal fading of TL signal often attracted attention of the researchers [4-35]. The usual method for studying the thermal fading is to irradiate the samples, at a constant temperature, and then to read out the samples atdifferent time intervals after storage in constant temperature conditions.

The aim of the present chapter is to study the influence of temperature and duration of storage after irradiation on the thermal fading of the TL signal. However, it is noted that, most of the TLDs exhibit more than one glow peak [3]. For example, the glow curve of TLD-100 has been reported to have up to 13 glow peaks from room temperature to 400 °C [8,36]. The depth (in terms of energy) of these glow peaks is not always increases by increasing the heating temperature. For example, in case of TLD-100, the trap depth of the glow peak appearing at 190 °C equal to 2.2 eV, whereas the glow peak appearing at 260 °C having activation energy equal to 1.8 eV [8]. Then it became important to know for how long is the dependence of the thermal fading on the trap depth for every glow peak, separately. Also, the dependence of the thermal fading on the frequency factor was not considered before. Effect of the order of kinetics on the remaining fraction of TL signal after long time of storage will be discussed.

The present work will discuss the dependence of the thermal fading on the absorbed dose in case of first-, second-, and general-order kinetics. The above mentioned studies will be given considering the models of first-, second- and general-order kinetics [37, 38, 39].

THEORETICAL BACKGROUND

2.1. First-order kinetics:

The starting model regarding to the TL phenomenon was pioneered by Randall and Wilkins [37]. They have considered the occurrence of a TL peak due to the thermal release of electrons from traps and their subsequent recombination with holes in recombination centers. The probability of electrons being retrapped is assumed in this model to be impossible. Therate of release of electrons per unit time at a temperature T (K) is given by:

$$-\frac{dn}{dt} = nS \exp\left(-\frac{E}{kT}\right) \qquad (1)$$

where n (cm-3) is the electron concentration at time t(s), E (eV) the depth of the trap level, S(s-1) the frequency factor and k (eV/K) the Boltzmann's constant. If the samples, after irradiation, are stored at constant temperature for lengths of time, then by rearranging equation (1):

$$-\frac{dn}{n} = S \exp\left(-\frac{E}{kT}\right) dt, \qquad (2)$$

where T is now a constant. This equation can be integrated to give the decay of the TL output with time at a given temperature as follows:

$$n(t) = n_0 \exp\left(-St \exp\left(-\frac{E}{kT}\right)\right), \qquad (3)$$

The equation describing TL for first-order is given by [9]:

$$I = n_0 S \exp\left(-\frac{E}{kT}\right) \exp\left[-\frac{S}{\beta}\int_{T_0}^{T} \exp(-E/kT^1) dT^1\right] \qquad (4)$$

where n_0 is initial concentration of trapped electrons at the starting heating temperature T_o and $\beta(Ks^{-1})$ the linear heating rate. This is the expression for the TL glow intensity I from electrons trapped at a single trapping level.

2.2. Second-order kinetics:

Garlick and Gibson [38], considered the case where electrons may be retrapped in addition to undergoing recombination with holes. In this model, the rate of release of electrons per unit time at a temperature T is given by:

$$-\frac{dn}{dt} = \frac{n^2}{N} S \exp\left(-\frac{E}{kT}\right) \qquad (5)$$

The decay of TL output in this case is given by the following equation:

$$n(t) = n_0 \left[1 + n_0\left(\frac{S}{N}\right) t \exp\left(-\frac{E}{kT}\right)\right]^{-1} \qquad (6)$$

where N (cm-3) is the concentration of traps. The equation describing TL for second-order is given by [38]:

2.3. General-order kinetics:

In several cases, the shape of the glow peak does not conform to either the first- or second-order glow curves expected from equations (1) and (5) respectively. Earlier authors [40-47] have suggested an equation for the general-order kinetics of an isolated TL glow peak which cannot be explained by the usual first- or second-order kinetics. The difficulty with respect to the old expression is related to the dimension of the pre-exponential factor which has unacceptable meaning. Rasheedy has improved a new expression for the general-order kinetics, which overcomes the previously mentioned difficulties [39]. The rate equation of this expression for the general-order kinetics is written in the following from:

$$-\frac{dn}{dt} = \frac{n^b}{N^{b-1}} S \exp\left(-\frac{E}{kT}\right). \qquad (8)$$

The decay of TL output in this case is given by the following equation:

$$n(t) = n_0 \left[1 + St\left(\frac{n_0}{N}\right)^{b-1}(b-1)\exp\left(-\frac{E}{kT}\right)\right]^{I/(I-b)} \qquad (9)$$

Where *b* is the order of kinetics, $1 \le b \le 2$.

Equation (9) includes the decay of the better known second-order case, *b*=2, given in equation (6). It is worthwhile noting that, the frequency factor S appearing in equation (9) has the dimension of (s^{-1}) similar to the frequency factor in case of the first- and second-order of kinetics without any difficulties related to the dimension problems. Putting b=2, equation (9) gives easily the decay of second-order glow peak (cf., equation (6)). Although equation (9) is not valid for the case b=1, it can easily shown that it reduces to equation (3) when b 1.

RESULTS AND DISCUSSION

3.1. Dependence of the thermal fading on the order of kinetics:

According to equations (3), (6) and (9) it is possible to calculate the TL response expected for dosimeters irradiated and stored for different intervals at a constant temperature. Figure (1) illustrates the fraction of TL signal remaining after storage at temperature 25 °C. Curve (a) depicts the TL fading for first-order according to equation (3), curve (b) for the general-order $b= 1.5$, by using equation (9), and curve (c) for the second-order kinetics, respectively. In figure (1) and for the sake of simplicity an intermediate value of the general-order kinetics $b=1.5$, was selected, whereas any other value of b between 1 and 2 may be used. This figure shows that the thermal fading increases in the direction of decreasing the order kinetics.
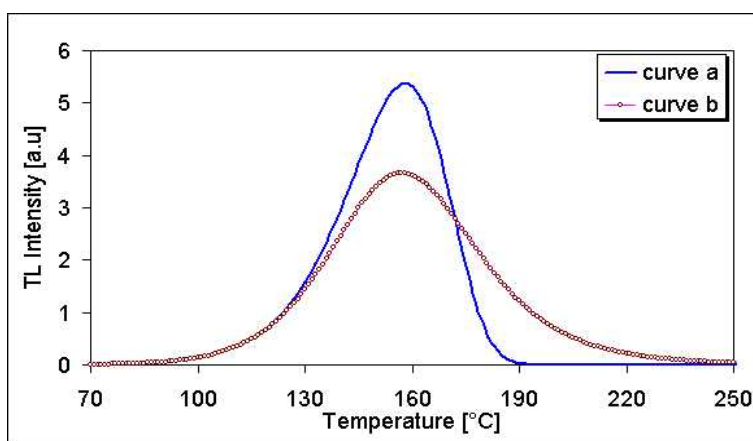
Figure (1): The fraction of TL signal remaining after storage temperature at 25 °C as a function of duration of storage. Curve (a) first-, (b) general- b=1.5, (c) second-order of kinetics. The curves are computed with E=1.1 eV and S=1x10^{12} s^{-1}.

Comparing with higher orders, the increase of the fading in case of first-order kinetics could be understood if one considered that in case of the first-order there is no chance of retrapping or trapping of the released electrons into other trap or recombination into other recombination centers [37]. This means that the electron immediately escape from its trap level will recombine rather quickly with the recombination center, while in case of second-order [38], the probability of retrapping increases as compared with the first-order case, which means a delay in the light emitted from the TL signal. This is the reason in front of the appearance of second-order glow peak broader than the first-order glow peak, with lower TL intensity comparing with the first-order glow peak sharing in the same trap parameters, while the total area of the two peaks is typically the same.Figure (2), depicts a computed first- and second-order TL glow peaksbased on equation (4) and (7), respectively. For this reason, and as a result of retrapping probability in case of second-order, a delay in the thermalfading of the TL signal with increasing the time of storage must beexpected.Then it might be reasonably expected that a first-order will display more fading than higher orders will do under the same conditions.
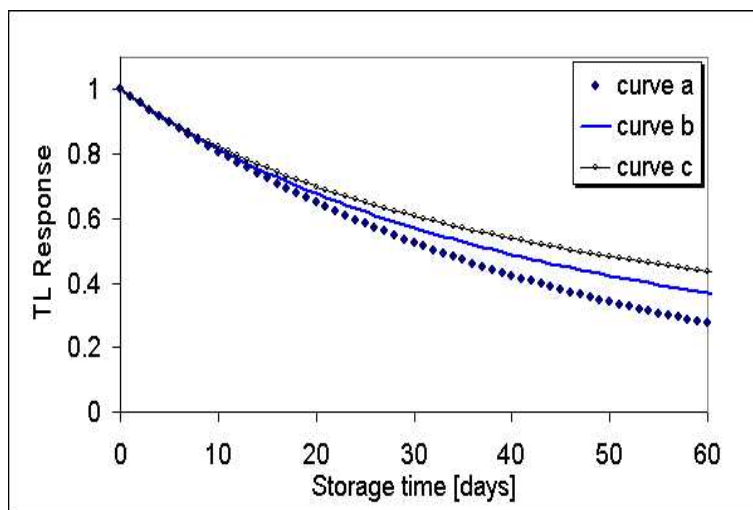
Figure (2): Computed TL glow peaks according to equations (4) and (7) of curve (a) first-order and curve (b) second-order of kinetics, respectively. The curves computed with E=1.1 eV, S=1x1012 s-1, no=N=104 cm-3 and =2 Ks-1.

3.2. Influence of temperature and duration of storage on the stability ofTL signal:

Figure (3) gives the TL response of first-order glow peak stored at 0,20, 30, 35 and 40 °C respectively. These curves are calculated by usingequation (3). Generally, it is seen from these curves that the fading increases with increasing the period and the temperature of storage.
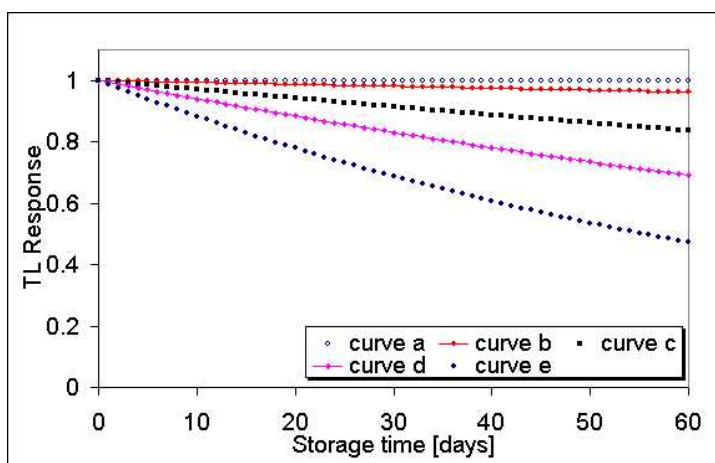

Figure (3): The fraction of first-order TL signal versus storage time.
The temperature of storage are given as a=0, b=20, c=30, d= 35 and e=40 °C.
The curves are computed with E=1.17 eV and S=3x10^{12} s^{-1}.

It can be seen from these curves that while low temperature storage may result in negligible fading, storage under elevated temperature conditions can result in significantly higher loss of signal depending on the characteristics of the trap parameters of the glow peak. The effect of trap parameters E and S will be discussed in the following subsection.

3.3. Effect of the trap depth and the frequency factor on the TL response of the glow peaks:

In addition to the order of kinetics, each glow peak is characterized by two parameters, the depth of trap level E and the frequency factor S. Figure (4) shows the dependence of TL response on the depth E of several first-order glow peaks, calculated according to equation (3). All of these glow peaks having the same value of frequency factor S. The assumption also is that all of these samples have been kept for intervals expanded to 60 days at constant temperature 27 °C.
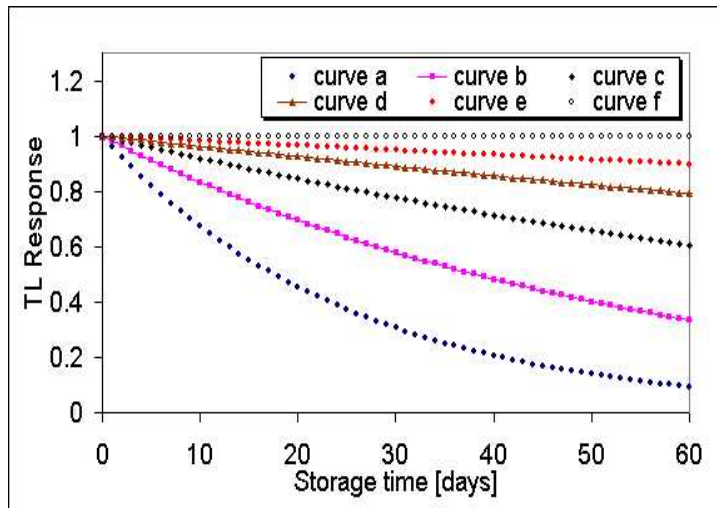
Figure (4): Fading of several first-order glow peaks having the same frequency factor S=3x10^{13} s^{-1}, with different values of the activation energy *E*, a=1.12, b=1.14, c=1.16, d=1.18, e=1.2 and f=1.3 eV.

On the other hand, figure (5) represents several glow peaks having the same trap level E=1.2 eV with different values of the frequency factor S, have been kept under the same conditions of figure (4).
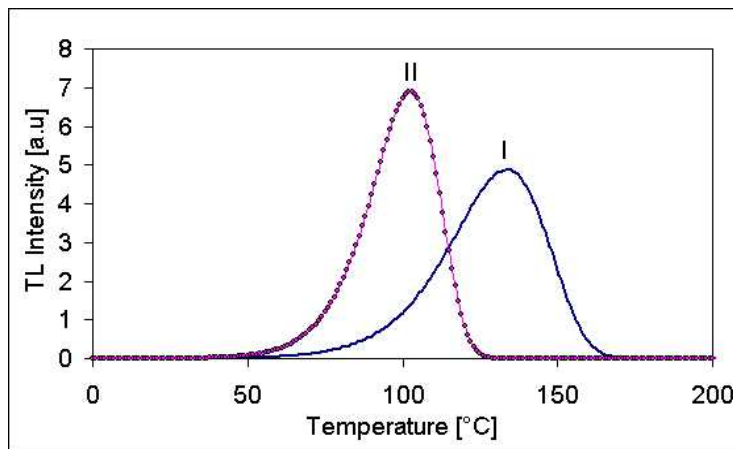
Figure (5): Fading of several first-order glow peaks having the same activation energy E=1.2 eV with different values of the frequency factor S, a=1x1011, b=7x1012, c=2x1013, d=5x1013 and e=1x1014 s-1.

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It is clear from figures (4) and (5) that the response of TL signals decreases in the direction of decreasing the trap depth and of increasing the frequency factor. This means that not only the depth of the trap level acts as a parameter controlling the thermal fading, but also the frequency factor which plays an important role in the fading of TL signal.

In case of glow curve including more than one TL glow peak one may expect that the higher temperature glow peak would be thermally more stable than the lower temperature peak. Unfortunately, this is not always true. The following example serves as a good illustration of the importance of the relative of S and *E*. Consider two first order traps with E_1=0.880 eV,S_1=1x10^{10} s^{-1}, E_2=1.077 eV and S2=5x10^{13} s^{-1}, respectively. At a heating rate =2 Ks-1, the TL glow peak according to E_1 and S_1 would occur at 135 °C in a glow curve, while the other glow peak according to E_2 and S_2 would be at 104 °C as shown in figure (6). The glow peaks are calculated according to equation (4).
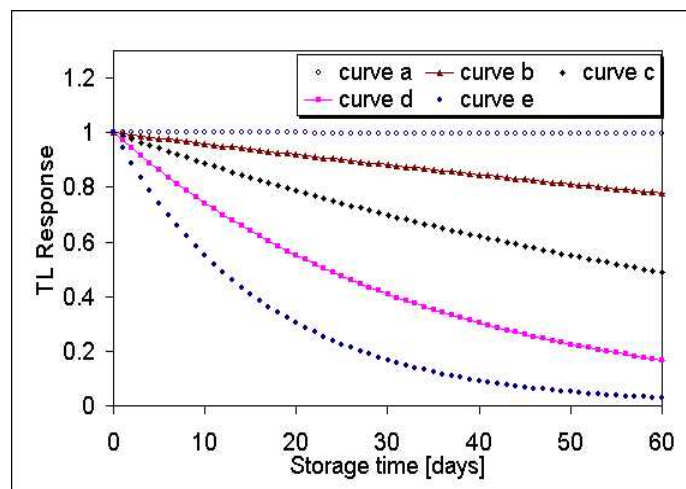
Figure (6): First-order glow peaks computed with (I) *E*=0.880 eV and S=1x1010 s-1 and (II) *E*=1.077 eV and S=5x10^{13} s^{-1}.

If the samples are stored at -5.1 °C, the fading curves of the two peaks would coincidence as shown in curve (a), figure (7). However, if the storage temperature is increased to 5 °C the low-temperature glow peak at 104 °C would decay faster than the higher one as shown in curve (b) and (c) of figure (7), respectively. Conversely, if the storage temperature was decreased to -12 °C, the high-temperature glow peak at 135 °C will actually decay faster than the lower temperature glow peak at 104 °C, as shown in curve (d) and (e) of figure (7), respectively. Thus, it must not simply be assumed that the fastest decaying signal belongs to the lowest temperature glow peak.

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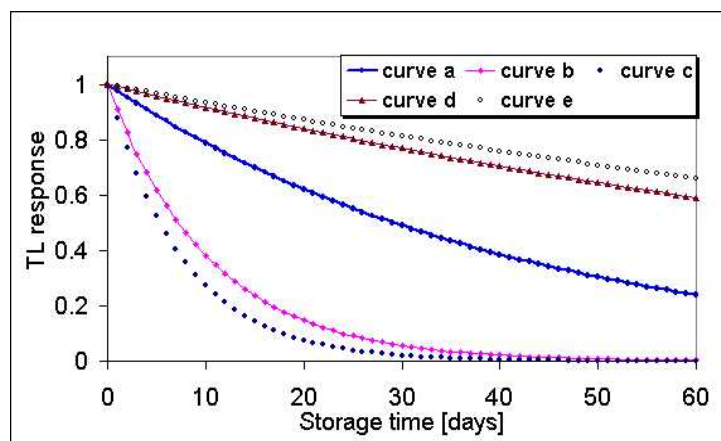
Figure (7): Calculated the fraction of TL signal versus storage time for the two glow peaks appearing in figure (6).The temperature of storage in curve (a) at -5.1 °C, (b) and (c) at 5 °C and (d) and (e) at -12 °C. Curve (a) represents the fading for either of the two peaks while curves (b) and (d) represent the fading to peak I and curves (c) and (e) represent the fading due to peak II.

On the other hand, it was found that several materials show that the lowest glow peak having the higher values of the trap depth E [8, 48-52]. For example, Hubner et. al. [50] has found that in case of CaF2:Mn the trap depth and frequency factor of the glow peak appearing at 212 °C are 1.2 e and 1011 s-1, respectively and of the glow peak appearing at 275 °C are 0.92 eV and 107 s-1, respectively. Both of them are first-order kinetics. According to equation (3), the thermal fading at any storage temperature from the glow peak appearing at 275 °C is much higher than the low temperature glow peak at 212 °C.

According to these calculations, one should not expect that the higher temperature glow peak is the most stable one. In other words, the depth of trap level of a glow peak is not the unique parameter to estimate the stability of this peak, but there is also another parameter, the frequency factor which acts in the opposite direction with respect to the depth of the trap level.

Studies on the effect of the depth of the trap level and the frequency factor on TL glow intensity show that thermal fading from deeper trap level could occur stronger than shallow trap level depending on the frequency factor of both of these glow peaks as well as on the temperature of storage. Consequently, when dosimeters susceptible to temperature dependent effects are used, then attention should be given to the trap parameters of the glow peaks.

3.4. Influence of the absorbed dose on fading of TL signal.

Figures (8)-(10) display the dependence of thermal fading on the absorbed dose in case of first-, second- and general- order kinetics, respectively. The absorbed dose is proportional to the initial concentration of trapped electrons no. In these figures, the temperature of storage is kept constant at 20 °C. Equations (3), (6) and (9) are used to calculate the TL response of the first-, second- and general-order kinetics, respectively.

It is clear from these two figures that, the thermal fading is independent parameter of the absorbed dose in the case of first-order, while in case of second- and general-order the thermal fading increases by increasing the concentration of trapped electrons. This theoretical data conform with experimental results previously been reported by different authors [53, 54].

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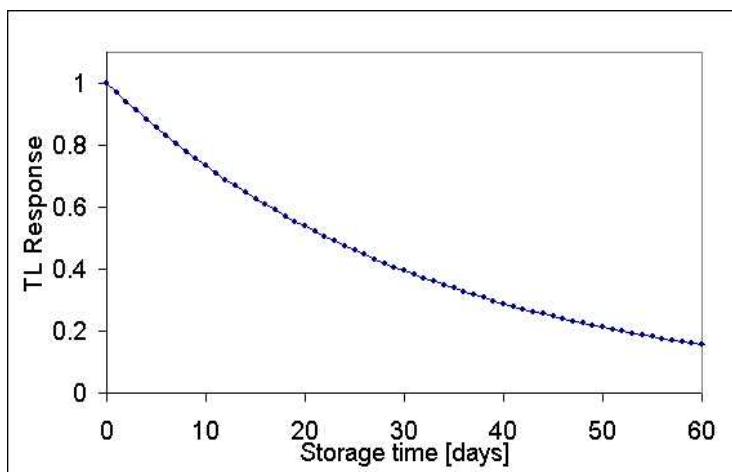
Figure (8): Calculated the fraction of first-order TL signal versus storage time. The curve is adequate for any initial number of trapped electrons. The curve is computed with E=1.1 eV and S=3x10^{12} s^{-1}. The temperature of storage is kept constant at 20 °C.

The fading is dose independent in case of first-order, and the fading in case of second- and general-order kinetics is found to be dose dependent and is higher at high doses. This observation may be interpreted on the following words. Let us consider two traps having the same parameters E and S. One of them is first- and the other one is second-order. Both of them are irradiated with the same dose, i.e., having the same no. The difference between the two peaks is only related to the probability of retrapping of the released electrons.
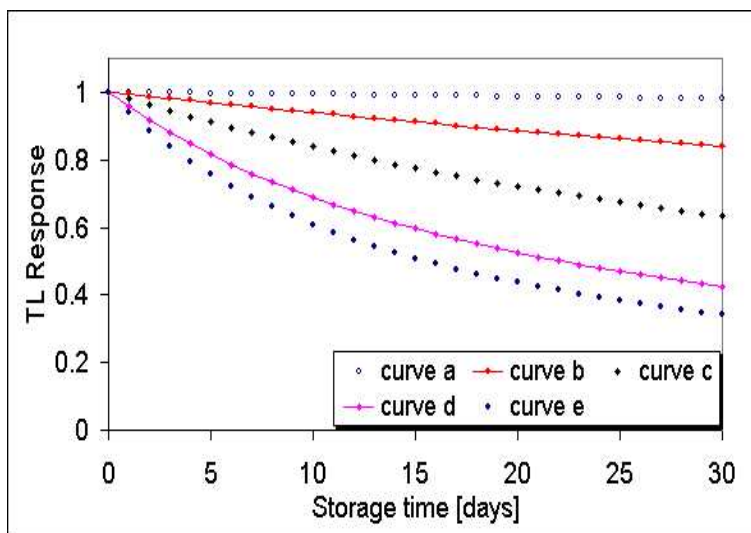
Figure (9): Calculated the fraction of second-order TL signal versus storage time. The initial number of trapped electrons are given a=1x10^2, b=1x103 c=3x10^3, d=7x10^3 and e=1x10^4 cm^{-3}. The curve are computed with E=1.1 eV, S=3x10^{12} s^{-1} and N=1x10^4 cm^{-3}. The temperature of storage is kept constant at 20 °C.

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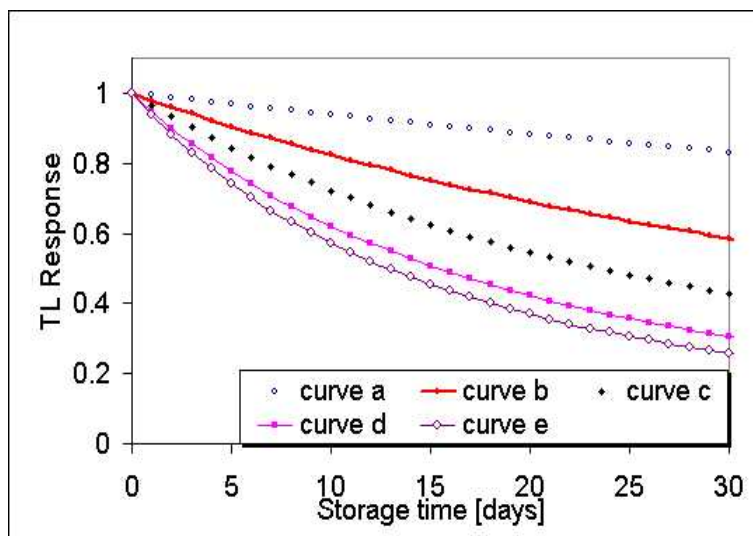
Figure (10): Same as figure (9) in case of general-orde TL signal with b=1.5.

In case of first-order, there is no chance of retrapping. Therefore, the electrons escape from the trap level will immediately recombine with holes in the recombination centers, giving rise of TL emission. This means that the rate of fading is independent parameter of the initial concentration of trap electrons and is not correlated with the concentration of trap centers. In the case of the second-order, we shall consider two cases. Firstly, when the concentration of trapped electrons no is much less than the concentration of traps N. In this case there is a high probability of retrapping, consequently a less of fading. Secondly, by increasing of the absorbed dose, i.e., increasing the concentration of trapped electrons, the last probability of retrapping decreases gradually and the probability of recombination with opposite centers gradually increases, giving rise of much fading at the higher doses.

CONCLUSIONS

It is found that the thermal fading at constant temperature increases with increasing the time of storage after irradiation. The results indicate that while low temperature storage may result in negligible fading, continuous storage under elevated temperature conditions can result in significantly higher loss of signal depending on the trap characteristics.

The fading at a fixed temperature increases with decreasing the order of kinetics. However, studies on the effect of the depth of the trap level and the frequency factor on TL glow intensity show that thermal fading from deeper trap level could occur stronger than shallow trap level if the former has lower value of the frequency factor than the latter. The results indicate that the thermal fading independent of the absorbed dose in case of first-order kinetics, while in case of second-order the thermal fading is found to dose dependent and the fading increase by increasing the absorbed dose.

Thus, attention should be given to the trap parameters of the glow peaks when dosimeters susceptible to temperature and time dependent effects are used.

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