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HEAVY ION RADIATION DAMAGE ANNEALING MODELS—A NEW INTERPRETATION

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The annealing behaviour of heavy ion radiation damage trails in Lexan Polycarbonate is studied on the basis of two different models proposed by Modgil and Virk[11] and Price *et al.*[15] The shortcomings of these two approaches are brought into focus and a new formulation proposed to remove some inherent drawbacks.

Key words: damage, heavy ions, polymers, annealing.

1 INTRODUCTION

The passage of a nuclear particle through a solid state track detector (SSTD) leaves behind its footprints in the form of a damage trail which holds crucial information for analysis about the particle and its interactions in the medium. Immense efforts have been made for the collection of such data. Some useful information can be derived if we demolish the footprints of recorded particles; the process is known as annealing. The annealing mechanism has been mainly studied in minerals and glasses due to its extensive application in fission track dating.[1-6] Most of the authors consider the damaged region to be constituted of many defects aggregated to one single defect. The kinetics of annealing is described by an Arrhenius type relation

$$t = a \exp(E/KT) \tag{1}$$

which predicts the time-temperature (t, T) combination necessary to achieve a particular degree of annealing. Equation (1) does not allow treatment of the problem of annealing for a given temperature history $T(t)$. To overcome this difficulty various authors[7-12] have presented different empirical relations. Dakowski *et al.*[7] proposed a linear relationship between the extent of track annealing and ln t for tracks in mineral grains annealed isothermally. Roberts *et al.*[3] introduced the concept of the energy per etchable defect for SSTRs. Mark and Mark[8] have described the isothermal fission track annealing as a diffusion process in terms of an infinite series of exponential decay functions. Haack[9] considers the temperature history to be made up of a finite number of isotherms over suitable time intervals. Recently, the gap model of Dartyge *et al.*[10] considering the latent track to be constituted of point and extended defects has acclaimed wide popularity. However Modgil and Virk[11] have adopted a more realistic approach by introducing the concept of annealing rate in their empirical formulation to understand and interpret the annealing behaviour in SSTDs.

There are only a few reports on annealing in plastic track detectors. It is true that the complex nature of plastics makes it difficult to present an explanation of the annealing mechanism. Single Activation Energy model[11] was found to give a best fit to data collected for CR-39 plastic track detector.[13] This has been confirmed by

Salamon *et al.*[14] Later on, Price *et al.*[15] introduced a modified version of Modgil and Virks equation by substituting annealing rate with reduced etch rate to explain their annealing data on phosphate glasses.

The authors state: "Using this surprisingly simple formula, we can predict the thermal fading rate as a function of annealing time and temperature for any ion at any velocity in the interval tested. We can then easily determine the extent of degradation of charge resolution for all species of interest, caused by differences in thermal history of old and young tracks".

In this paper we present a comparison of the models proposed by Modgil and Virk[11] and Price *et al.*[15] and evolve a new approach to interpret the annealing data in Lexan Polycarbonate.

2 ANALYSIS OF ANNEALING MODELS

Samples of Lexan Polycarbonate were irradiated with heavy ion beams of 14.6 MeV/n ^{139}La, 13.6 MeV/n ^{208}Pb and 16.0 MeV/n ^{238}U from the accelerator facility at GSI, Darmstadt. These samples were annealed between 100–160°C for time intervals of 10, 20, 40 and 60 min, respectively. The corresponding lengths of both the unannealed and annealed samples were recorded after etching the different sets of samples with 6 N NaOH at 60°C. These were then used for analysis of the various models. We have projected here curves drawn for only ^{238}U ion beam, whereas the results for ^{208}Pb and ^{139}La have been tabulated, as the curves drawn for these ions show correspondence with those of ^{238}U.

2.1 *The Annealing Model of Modgil and Virk*

The three-step annealing model presented by Modgil and Virk was successfully applied to various inorganic track detectors. The authors predicted that the activation energy of annealing, E_a, was the property of the bulk material, and hence a constant. They proposed an empirical formula

$$V_a = A\, t_a^{-n} \exp(-E_a/KT), \tag{2}$$

where $V_a = dl/dt$ is the annealing rate, t_a and T_a are the annealing time and temperature, respectively, n, the exponent of t_a and A, the proportionality constant. When this formula was applied to Cr-39 data, for the first time by Virk *et al.*[13] it was realized that the formulation had a number of drawbacks; for instance, although the annealed length observed for different degrees of annealing had to be a function of the track etch rate, V_t, the effect of this parameter was neglected. This is obvious from the fact that in their methodology, the authors propose an etching time t_c (the time required for the full range of the unannealed tracks to be revealed) for the etching of both the unannealed and annealed samples; which implies that instead of the true annealed lengths being observed, one obtained reduced lengths of the annealed tracks. To remove this drawback it is reasonable to assume that if we select the etching time to be small, we would be projecting the effect of etch rate, V_t, in terms of length. In our present study, we have chosen the etching times for ^{208}Pb, ^{238}U and ^{139}La ions to be 45, 30 and 30 min respectively. This choice is necessitated by the fact that the linear portions of all annealed curves should be crossed by a line drawn parallel to the y-axis from the corresponding etching time.

The instantaneous annealing rates are obtained from the plot between these annealed lengths and their respective annealing times. These rates are expressed in terms of Eq. (2) to compute values for E_a and n. It is observed that E_a and n are independent of the extent of annealing. Moreover, E_a is also independent of the type and energy of ion beam used, confirming the hypothesis of single activation energy proposed by Modgil and Virk.[11] The values of E_a and n for all the ions are tabulated in Table I.

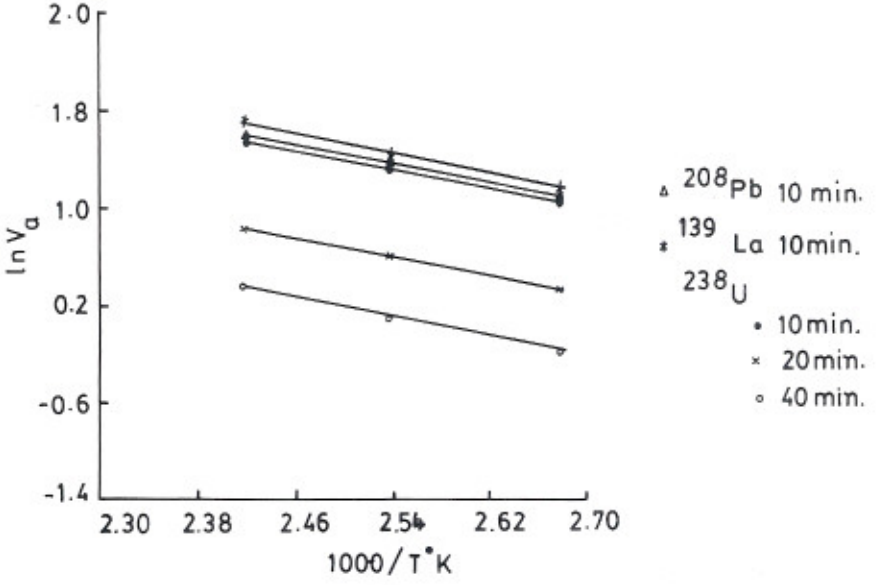

Figure 1

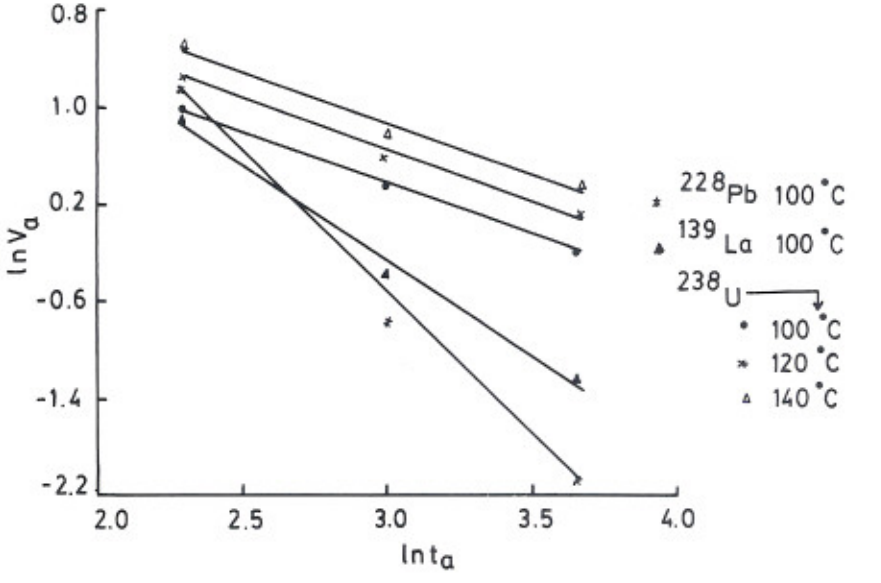

Figure 2

TABLE I
The values of activation energy E_a and exponent n in Lexan Polycarbonate using different formulations

	Modgil & Virk		Price *et al.*		New formulation	
Ion beam	E_a(eV)	n	E_a(eV)	n	E_a(eV)	n
^{238}U	0.171	0.847	0.169	0.895	0.170	2.625
^{208}Pb	0.173	2.40	0.170	0.935	0.171	3.00
^{139}La	0.173	1.54	0.172	0.913	0.170	2.14

2.2 *Formulation of Price et al.: The Modified Version*

In a recent paper on phosphate glasses, Price and co-workers[15] found that their annealing data could fit into the equation of Modgil and Virk provided the thermal-fading rate is replaced by the fractional thermal-fading rate

$$\frac{(S-1)_i-(S-1)_f}{(S-1)_i}=A\,t_a^{1-n}\exp(-E_a/KT) \tag{3}$$

where $S=V_t/V_g$, and the subscripts i, f refer to initial and final values. The authors have further proposed that using this modified version of Eq. (2) the values of parameters A, n and E_a are reduced to certain constants of the detectors, irrespective of the ion beam used.

We took up the challenge of testing this formulation on our recent data on annealing of heavy ion tracks in Lexan Polycarbonate. We observed that the bulk etch rate, V_g, is independent of the extent of annealing and hence can be treated as a constant. The graphs (Figures 3, 4) plotted between the L.H.S. of Eq. (3) and $1/T$ and t_a, respectively, provide us with values of E_a, n and hence A for ^{238}U ions. The values for ^{208}Pb and ^{139}La ions have also been incorporated in Table I.

2.3 *A New Interpretation*

Using the formulation of Price *et al.*[15] it is observed that the fitting of curves is rather poor. Similar results are obtained for ^{208}Pb and ^{139}La ions. The authors themselves have commented[15]:

"Attempts to fit the fractional thermal annealing to functions of time other than a power law gave much worse results."

The values for n and E_a obtained using Eq. (3) indicate that E_a is an ion independent parameter, whereas n and hence A are found to depend on the ion beams used; a result in contradiction to that of Price *et al.*[15] This drawback was earlier confirmed and reported in our communication on CR-39 plastic.[16]

An attempt has been made to introduce a new parameter, instantaneous track etch velocity on the L.H.S. of Eq. (2) to remove the drawbacks discussed above and in Section 2.1. Our formulation

$$d/dt_a(V_t)=A\,t_a^{-n}\exp(-E_a/KT), \tag{4}$$

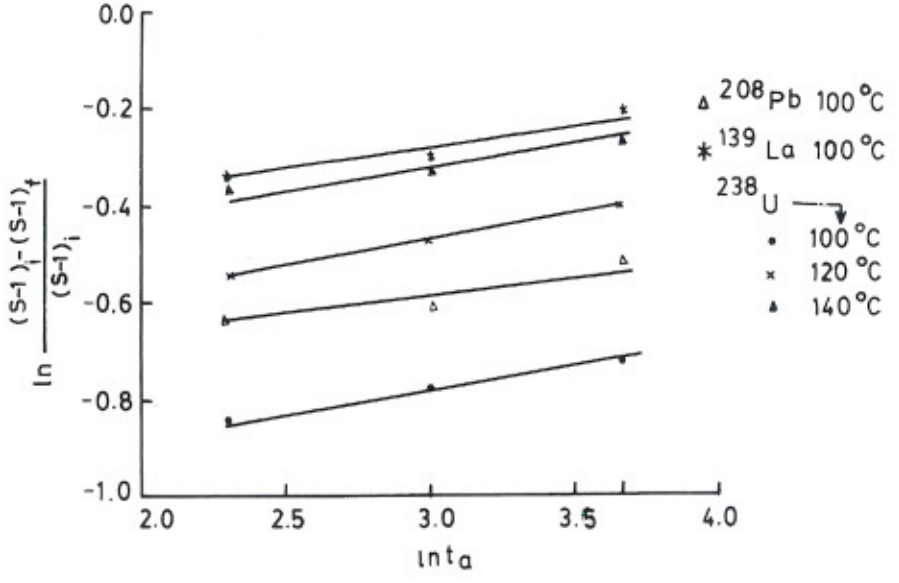

Figure 3

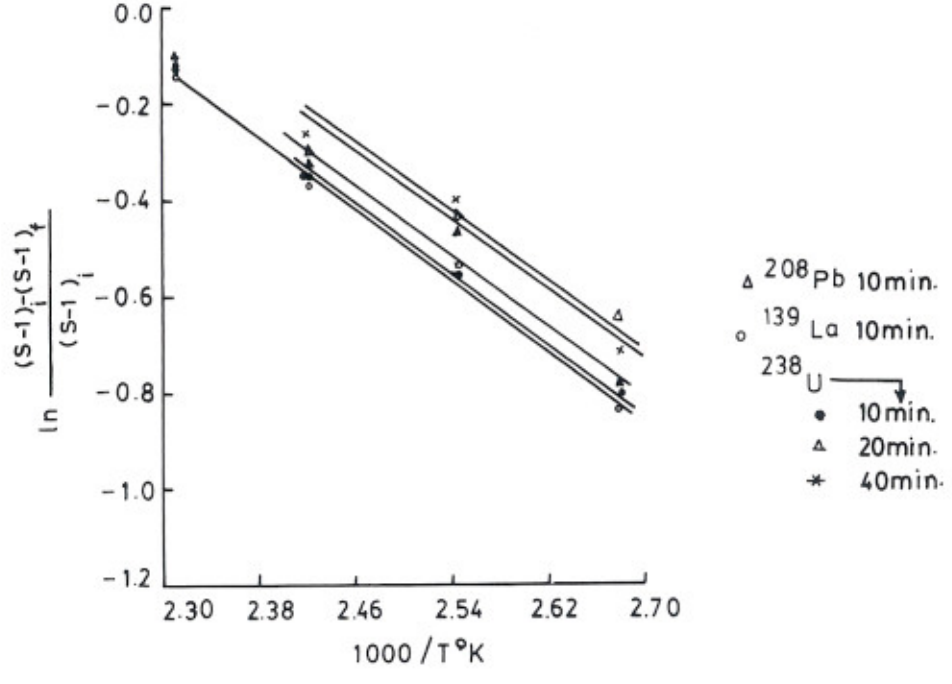

Figure 4

where $V_t = dl/dt_e$ is the etch rate at a particular annealing temperature and time; is found to give a better fit (Figures 5, 6) than the earlier projection by Price *et al.* It also removes the short-comings of Modgil and Virk's approach. Furthermore, the parameters n and A are functions of the linear energy transfer (as observed in other formulations) of the penetrating ion, whereas, E_a, activation energy of annealing turns out to be a property of the detector.

CONCLUSION

We have seen that the thermal fading of ^{208}Pb, ^{139}La and ^{238}U in Lexan Polycarbonate can be represented by a few, equivalent, empirical relations, perhaps the most useful of which is

$$d/dt_a(V_t) = A\, t_a^{-n} \exp(-E_a/KT)$$

The above formulation gives a more direct measure of annealing than either V_a or $[\langle S\text{-}I\rangle_i - \langle S\text{-}I\rangle_f]/\langle S\text{-}I\rangle_i$ which are more involved functions. From this new formulation one can infer that annealing may occur at even room temperatures provided the samples are stored for longer periods. This obviously affects experiments which use Lexan-polycarbonate for particle identification over an extended collection time.

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