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RADIATION DAMAGE ANNEALING MODELS IN GLASS DETECTORS

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The thermal annealing behaviour of heavy ions and fission fragment tracks in sodalime, phosphate, and silicate glass is studied. A review on thermal annealing formula is given. A simplified empirical formula is derived, comprising both isochronal as well as isothermal annealing procedures.

Key words: Heavy ion tracks, fission fragment tracks, isochronal annealing, isothermal annealing.

1 INTRODUCTION

Among the Solid State Nuclear Track Detectors (SSNTDs), glass detectors play a special role in heavy-ion experiments due to their higher registration threshold where it is desirable to eliminate all the backgrounds of light charged particles. A number of different glass detectors were examined by several authors^{1–6} for their suitability as nuclear track detectors.

Although the pace of development is so fast, the phenomenon of thermal annealing of latent tracks now studied over two decades is still poorly understood, perhaps due to the lack of a satisfactory theory of track formation. Despite the existence of numerous investigations about the annealing behaviour of fission tracks in minerals and glasses there is no generally accepted description of the annealing kinetics. The most successful track formation models introduced to explain the thermal fading of tracks apply to minerals.^{7–13}

Naeser and Faul⁷ gave an explicit relation for track retention in apatite as a linearly decreasing function of $\log t$. It was later confirmed by Naeser *et al.*¹⁴ for epidote, by Haack and Potts¹⁵ for garnet and by Burchart *et al.*¹⁰ for apatite. Dakowski *et al.*¹⁶ noticed that the track retention lines on Arrhenius plots are true for various minerals but not for glasses and thus drew a clear distinction between crystalline substances and glasses.

Mark *et al.*¹¹ proposed that the annealing of fission tracks in apatite can be explained by a summation series of exponential decay functions. Gold *et al.*¹² have proposed a general reaction rate theory for the annealing process. Burchart *et al.*¹⁷ analysed the available experimental data on fission track annealing in apatite and found that taking track reduction density as proportional to the logarithm of time was most appropriate for fitting these data.

Dartyge *et al.*¹⁸ studied nuclear tracks in silicates by small angle X-ray scattering and concluded the tracks to be constituted of extended defects and point defects. Rather than being distributed in the peripheral areas around the core, they assumed the point defects to be located in gap zones separating the extended defects. The etchability as well as the annealing behaviour of tracks in their “gap” model is determined by the properties of the extended defects.

Cantelaube¹⁹ has tried to extend and extrapolate the Arrhenius law to the case when fading temperature varies over time. Bertal and Mark¹³ studied the annealing

kinetics of fission tracks as a sum of exponential decay functions and concluded that the etchability of fission tracks is due to extended defects, i.e. systems of dislocation loops.

Modgil and Virk²⁰ introduced a single activation energy model to explain the annealing kinetics of fission fragments in sodalime glass. This model corresponds to the diffusional motion of displaced atoms during track formation back to their proper lattice positions.

Green *et al.*²¹ presented a qualitative description of fission track annealing by paying particular attention to the variation of confined track lengths during annealing. A mathematical description is formulated which supports the assumption of a single activation energy based on the results of annealing experiments on apatite.

Price *et al.*²² postulated an empirical relation with some modification in Modgil and Virk formula.²⁰ The relation supported the concept of single activation energy of annealing in phosphate glass track detectors.

Finally, all the formulas have been investigated in detail confirming the single activation energy model with one minor modification (as discussed in Section III).

II MATHEMATICAL DESCRIPTION OF ANNEALING MODELS

Most of the authors^{7, 10, 15, 23} follow an Arrhenius type relation between annealing time and temperature which is generally expressed by a Boltzmann equation of the form

$$t = A \exp(E/kT), \quad (1)$$

where E is activation energy, k the Boltzmann constant and A is a material constant. The results obtained using Eq. (1) show a series of fanning lines yielding a spectrum of activation energies for different degrees of annealing.

Mark *et al.*,⁸ on the other hand, suggested that the annealing behaviour of track densities in apatite could be explained by a relationship of the form

$$p(t) = p(0) \exp(-\alpha t), \quad (2)$$

where $p(t)$ represents the track density at the annealing time t , and $p(0)$ is the original track density. In this case the decay constant (annealing coefficient) α is given by a sum of two exponentials (corresponding to two activation energies E_{01} and E_{02}).

$$\alpha(T) = \alpha_{01} \exp(-E_{01}/kT) + \alpha_{02} \exp(-E_{02}/kT) \quad (3)$$

where α_{01} and α_{02} are annealing constants and E_{01} and E_{02} are the activation energies of two different diffusion processes involved. However, for high temperature annealing, the data of Mark *et al.*⁸ could be approximated with a single exponential decay function which was confirmed by Bertal.²⁴ Thus Eq. (3) reduces to a simplified approximation

$$\alpha(T) = \alpha_0 \exp(-E/kT). \quad (4)$$

Using the above relation, Eq. (2) can be written as

$$\ln(-\ln r) = \ln \alpha_0 - E/kT, \quad (5)$$

where $r = p(t)/p(0)$ is the track density reduction ratio. The slope of the plot of left hand side of Eq. (5) against the reciprocal temperature yields the activation energy of the process, E .

Dakowski *et al.*¹⁶ reported the isothermal annealing data for various minerals and glasses by a relation of the form

$$p(t)/p(0) = a - b \ln t, \quad (6)$$

which is valid for values of $p(t)/p(0)$ between 5 and 95%.

Mantovani⁹ suggested a linear relationship for the annealing of fission tracks in muscovite,

$$p(t)/p(0) = a - bt. \quad (7)$$

Although it appears that the above three relations [Eq. (2), (6) and (7)] represent the annealing data sufficiently well, Eq. (6) gave the minimum mean squared derivation of the annealing data for the best fit straight line¹⁷ which may be an accidental coincidence.

Green *et al.*²¹ postulated a model based on the track length measurements of confined tracks in apatite and described the experimental annealing data by best fit of the form

$$\ln(1-r) = C_0 + C_1 \ln t + C_2/T, \quad (8)$$

where $C_0 = 3.87$, $C_1 = 0.219$ and $C_2 = -4220.13$ are constants which seems to be a rather arbitrary choice for the functional dependence.

Laslett *et al.*²⁵ compared the logarithmic, exponential and linear models and embed them into a single "super model"

$$g(r) = \{[(1-r)^b/b]^a - 1\}/a = C_0 + C_1 \ln t + C_2/T, \quad (9)$$

$$\begin{aligned} \text{where } g(r) &= \ln(1-r) & \text{if } a=0, \quad b=1, \\ &= -r & \text{if } a=1, \quad b=1, \\ &= \ln(-\ln r) & \text{if } a=0, \quad b=0, \end{aligned}$$

but it has the disadvantage of a complicated structure and of being based on the assumption that the errors on the right hand side of Eq. (9) are normally distributed.

Modgil and Virk²⁰ proposed a three-step annealing model based on diffusion processes. The empirical formulation relating the annealing rate, V_a (defined as rate of change of length or diameter i.e. dl/dt or dD/dt) with activation energy, E is given as:

$$V_a = A t^{-n} \exp(-E/kT), \quad (10)$$

where A is a proportionality constant, n is the exponent of annealing time, t and the other symbols have their usual meanings.

Rewriting the above equation we have

$$\ln V_a = \ln A - n \ln t - E/kT. \quad (11)$$

The slope of the plot of $\ln V_a$ against inverse temperature will yield the activation energy E . The plot of $\ln V_a$ versus $\ln t$ will give the value of the exponent, n .

Price *et al.*²² introduced a modified version of above formulation in the case of phosphate glass detectors. Accordingly, the annealing data can be fitted well by replacing the thermal fading rate with a fractional thermal fading rate.

$$\frac{S_i - S_f}{(S - 1)_i t} = A t^{-n} \exp(-E/kT), \quad (12)$$

where $S = V_T/V_G$ and the subscripts i, f refer to the initial and final values; V_T , the track etch rate and V_G , the general etch rate.

Rewriting the above Eq. (12), we have

$$\ln \frac{S_i - S_f}{(S - 1)_i} = \ln A + (1 - n) \ln t - E/kT. \quad (13)$$

The slope of the plot of left hand side of Eq. (13) with inverse temperature will, as usual, yield the activation energy, E . However, it is observed that the general etch rate, V_G , in sodalime glass is independent of the extent of annealing and hence can be treated as constant, thereby making the fitting rather poor.

III MODIFIED BEST FIT MODEL

It is not possible to compare all these models here, but a systematic study yields an empirical formula which is much more simple and gives the best results. Considering the fitting errors as well as the simplicity of various models, it can be stated that the formula (10) combines the advantage of simplicity and reproduction of the experimental results.

Still, one has to realize that the formula (10) is not yet optimal. For example, (i) the empirical relation uses only an approximate derivative, i.e. $\Delta D/\Delta t$ and not the real one, dD/dt ; (ii) this formula yields $D - D_0$ while other formulae give D/D_0 ; (iii) moreover, the differential equations always have larger errors as compared with integral equations.

Therefore, an attempt is made to remove these drawbacks and improve formula (10). For obtaining a new formula, we modify Eq. (10), i.e.,

$$dD/dt = A t^{-n} \exp(-E/kT).$$

Integrating both sides, we get

$$\int_{D_0}^D dD/dt \cdot dt = A e^{-E/kT}, \quad \int_0^t \tau^{-n} \cdot d\tau,$$

$$D - D_0 = A \frac{t^{1-n}}{1-n} e^{-E/kT}, \quad (14)$$

where D_0 is the diameter of unannealed tracks. On simplification we have,

$$1 - r = A' \frac{t^{1-n}}{1-n} e^{-E/kT}, \quad (15)$$

where $r = D/D_0$, the track diameter reduction ratio, and $A' = -A/D_0$. We can rewrite Eq. (15) as

$$\ln(1 - r) = \ln A' + \ln(t^{1-n}/1 - n) - E/kT. \quad (16)$$

The plot of $\ln(1 - r)$ versus $1/T$ yields the activation energy E . The value of exponent n of annealing time t can be calculated from the relation

$$n = 1 - \ln(R_1/R_2)/\ln(t_1/t_2), \quad (17)$$

where $R = 1 - r$ and the subscripts 1 and 2 denote the values for different sets at constant temperature.

This relation (15) is also true for low cone angle glasses where $r = L/L_0$, the track length reduction ratio. Also, it is observed from the above relation (17) that the value of n will always be less than 1.

Thus, an improved formula is realized which removes the drawbacks of the original Modgil and Virk formulation [Eq. (10)] and merges with super model approach of Laslett *et al.*²⁵

This formula has been tested for annealing experiments in sodalime, phosphate and silicate glass detectors to corroborate the concept of a single activation energy in the annealing of radiation damage in glass as proposed earlier by Modgil and Virk.²⁰

IV EXPERIMENTAL DETAILS

The samples of sodalime glass were irradiated by ^{139}La ions of energy 14.6 MeV/u and ^{208}Pb ions of energies 17.0 and 13.6 MeV/u respectively, at GSI heavy ion accelerator, UNILAC. Samples of phosphate and silicate glasses are irradiated with a ^{252}Cf fission fragment source in a 2π geometry.

The irradiated samples of sodalime glasses were annealed in a Muffle furnace at temperatures of 100, 150, 200 and 250°C for time intervals of 10, 20, 40 and 80 min. All these samples, for each ion set, were etched for comparison simultaneously with the corresponding unannealed sample in 2.5% HF at room temperature (30°C) for 35 min. These etched samples were washed thoroughly in running water.

The irradiated samples of phosphate and silicate glasses were annealed isochronally at 400 to 650°C and 500 to 750°C with a 30 min holding time for each step of 50°C. The samples were etched in 40% HF, respectively, for 30 min and 5 min at room temperature (25°C) and then washed in water.

The track lengths and diameters are measured using a Carl Zeiss binocular microscope at a magnification of $1000\times$. The mean length, L_0 and diameter, D_0 of unannealed tracks is determined under identical conditions, as a reference standard. The track etch rate, V_T and the general etch rate, V_G , were also measured for calculating the reduced etch rate $S(=V_T/V_G)$ in sodalime glass samples irradiated with heavy ions. The track etch rate, V_T is calculated from the linear

portion of the plot of track length versus etching time. The general etch rate, V_G is confirmed to be independent of the annealing time and hence can be treated as a constant for sodalime glass. Only the track diameters of circular tracks corresponding to vertical incidence were counted in phosphate and silicate glasses.

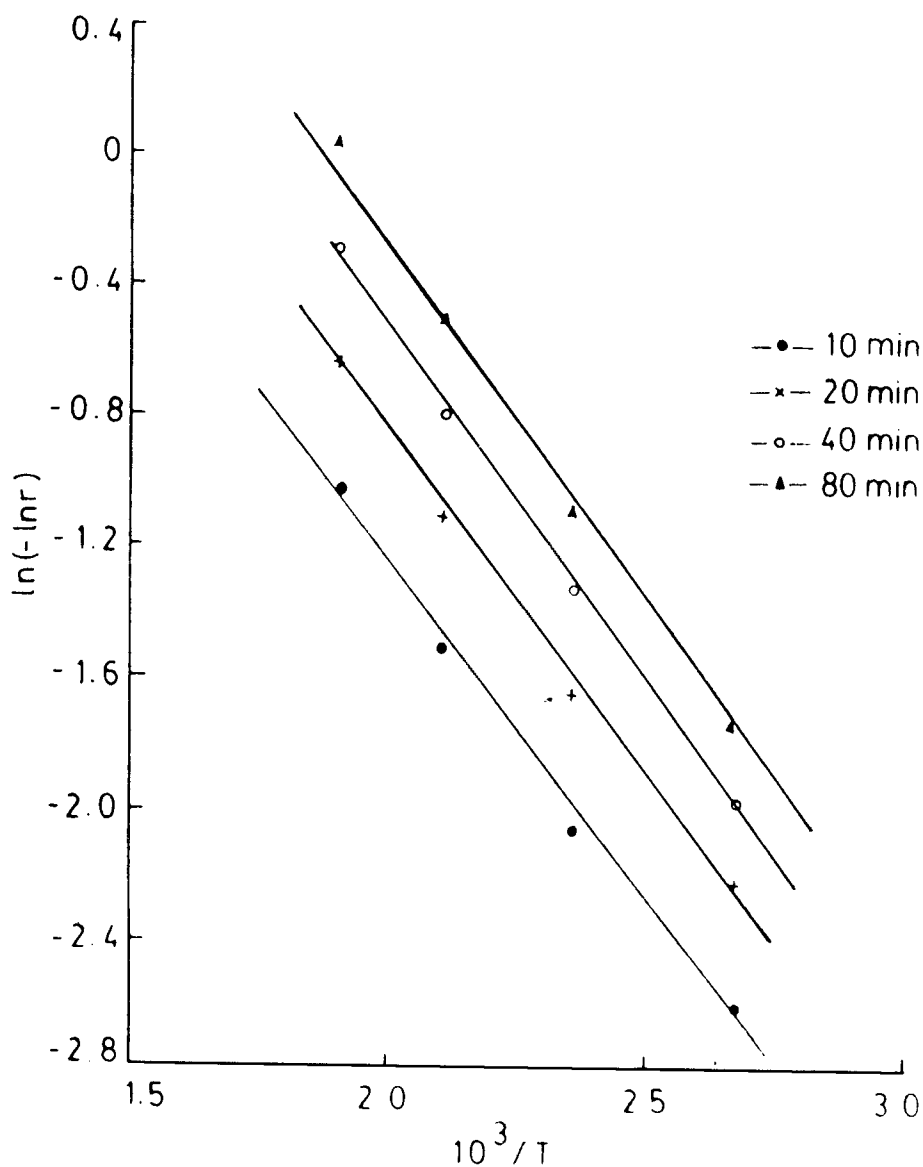


FIGURE 1 Plot of $\ln(-\ln r)$ vs. $10^3/T$ for sodalime glass using ^{139}La ions.

V DISCUSSION OF RESULTS AND CONCLUSIONS

We used the least squares method to fit the annealing data.

Figures 1-3 show the annealing results using Mark *et al.*, Modgil and Virk and Price *et al.* formulations in sodalime glass. The activation energies are calculated from the slopes of the plots of the left-hand sides of Eqs. (5), (11) and (13) against the inverse temperature in all the cases. The next plots (Figure 4) show the result of activation energy, E using our modified best fit approach according to Eq. (16). The resulting values of E and n in sodalime glass and their intercomparison using different formulas is given in Table I.

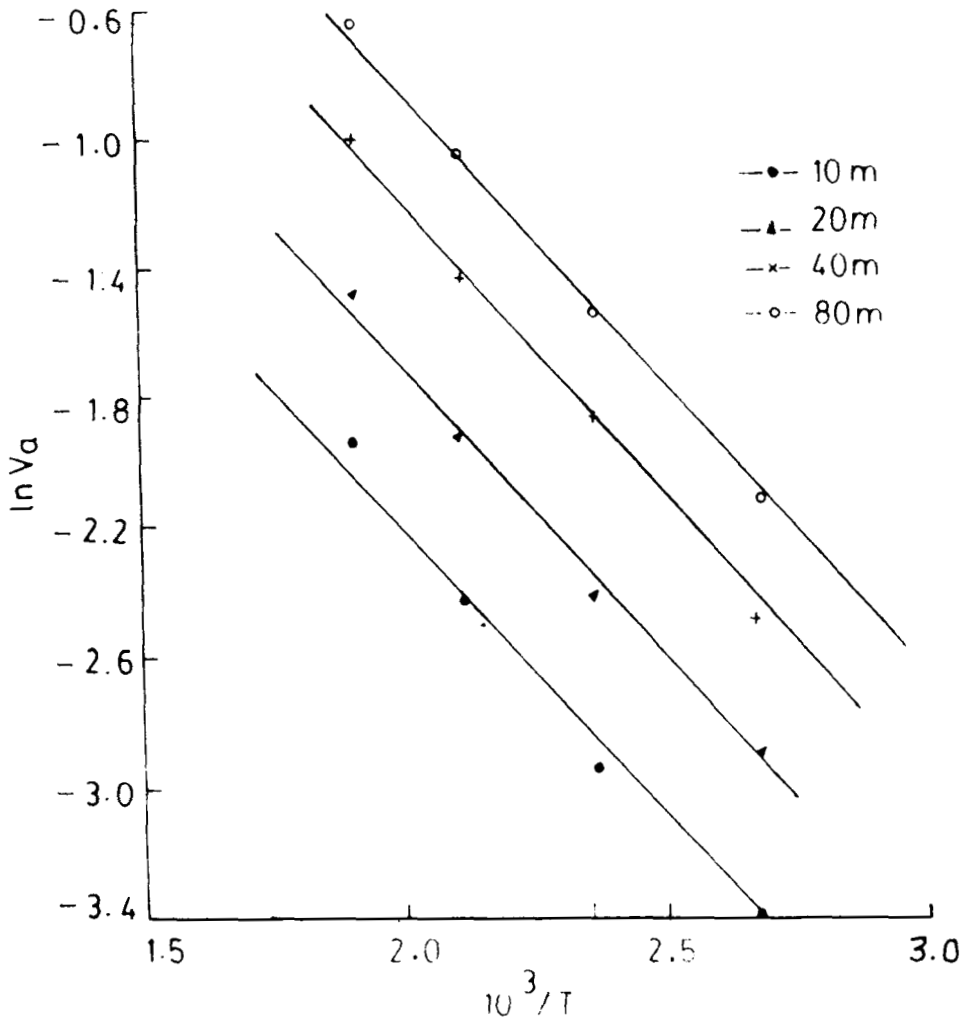


FIGURE 2 Plot of $\ln V_a$ vs. $10^3/T$ for sodalime glass using ^{139}La ions.

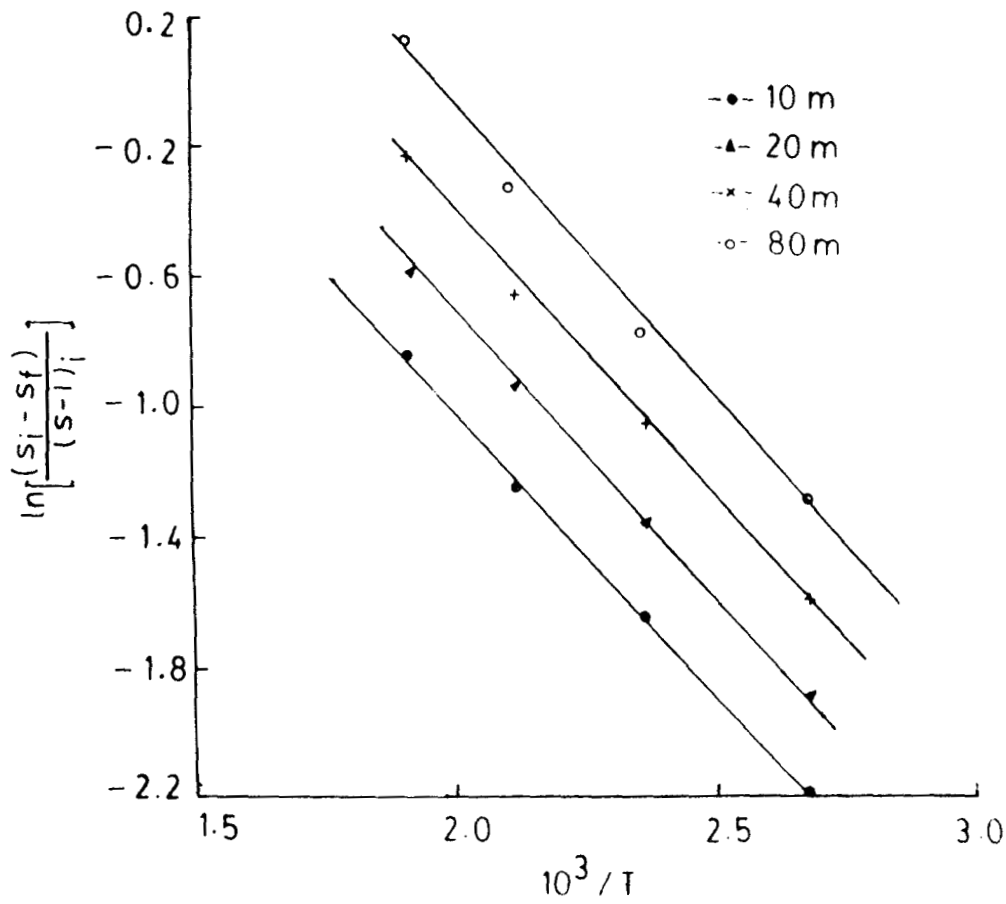


FIGURE 3 Plot of $\ln(S_i - S_f)/(S - 1)_i$ vs. $10^3/T$ for sodalime glass using ^{139}La ions.

TABLE I
Intercomparison of various formulations using sodalime glass as track detector

Ion beams used (MeV/u)	Mark <i>et al.</i>		Modgil and Virk		Price <i>et al.</i>		Modified approach	
	<i>E</i> (eV)	<i>n</i>	<i>E</i> (eV)	<i>n</i>	<i>E</i> (eV)	<i>n</i>	<i>E</i> (eV)	<i>n</i>
^{139}La (14.6)	0.18	0.45	0.16	0.64	0.15	0.55	0.16	0.64
^{208}Pb (17.0)	0.18	0.50	0.16	0.65	0.15	0.57	0.16	0.64
^{208}Pb (13.6)	0.18	0.40	0.16	0.60	0.15	0.52	0.16	0.64

The results of fission fragment tracks for isochronal annealing in phosphate and silicate glasses are calculated using our modified approach (Figures 5 and 6). The resulting activation energies for phosphate and silicate glass are 0.52 and 0.57 eV respectively.

The results obtained from our modified approach are quite satisfactory. However, the difference is not yet significant enough to justify the acceptance or rejection of one of the alternative approaches.

Therefore, we may conclude that while constructing empirical models to be used as a basis of prediction, the simple models with less fitted parameters are generally preferable. The model supports the concept of a single activation energy of annealing and removes the ambiguities of earlier models using the Arrhenius approach.

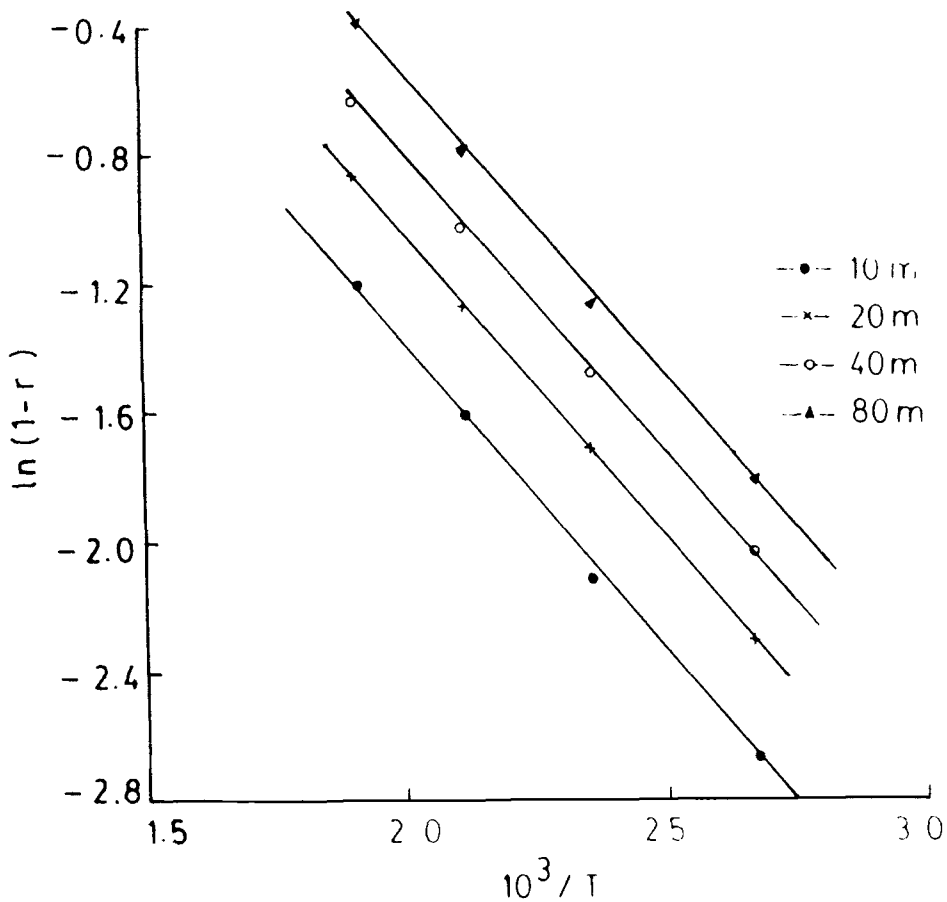


FIGURE 4 Plot of $\ln(1-r)$ vs. $10^3/T$ for sodalime glass using ^{139}La ions.

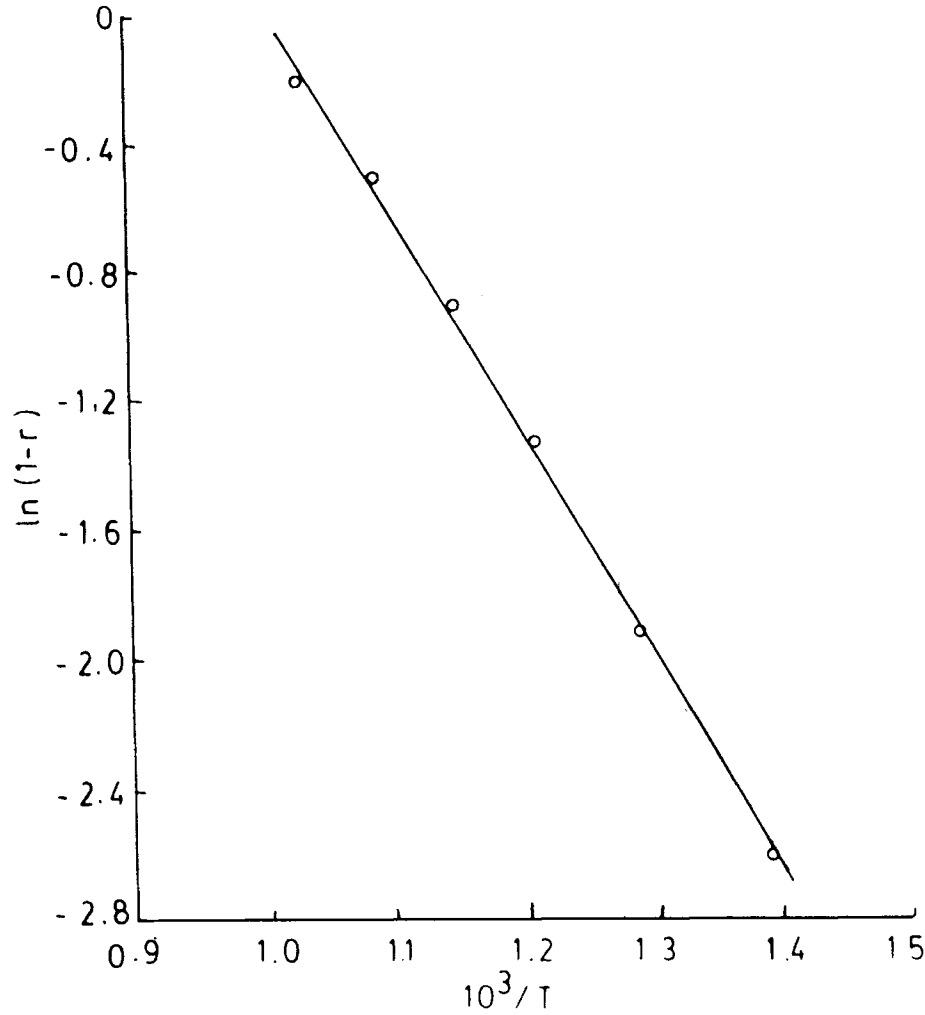


FIGURE 5 Plot of $\ln(1-r)$ vs. $10^3/T$ for phosphate glass using fission fragments.

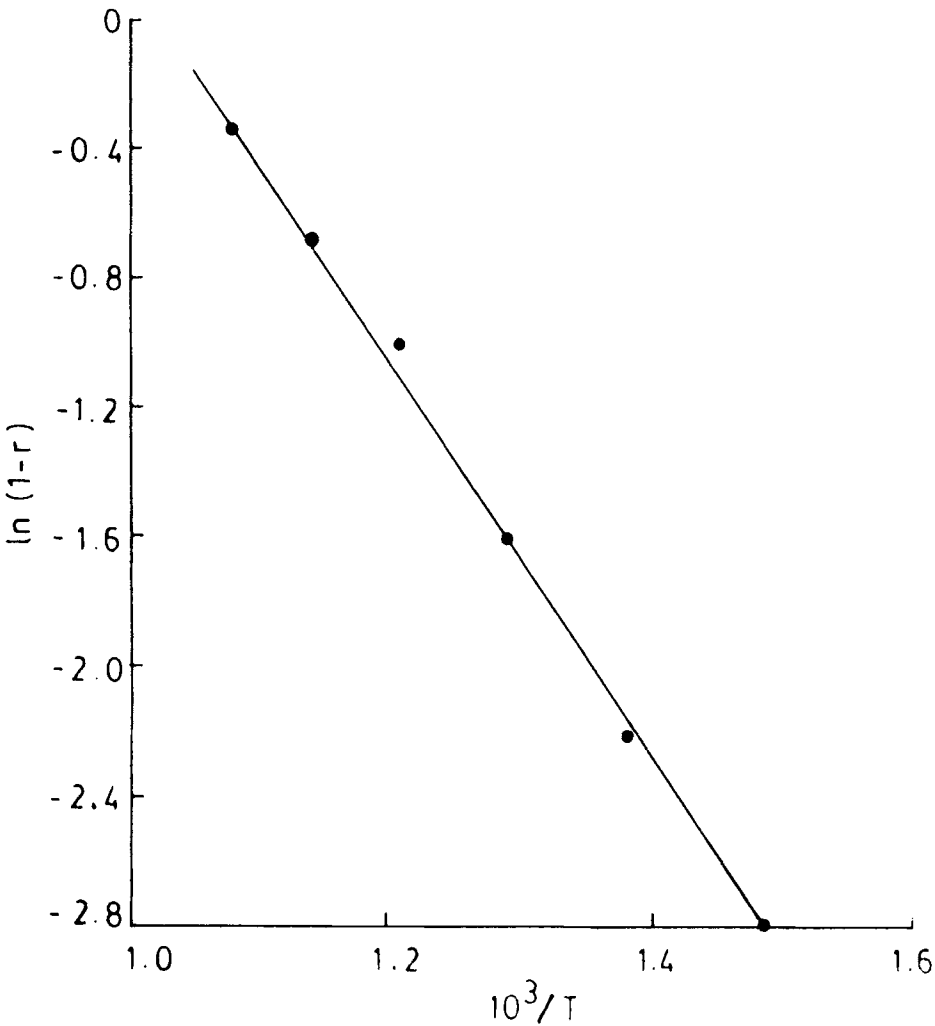


FIGURE 6 Plot of $\ln(1-r)$ vs. $10^3/T$ for silicate glass using fission fragments.

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