ANNEALING CHARACTERISTICS OF NUCLEAR TRACKS IN GLASS DETECTORS USING OPTICAL ABSORPTION SPECTROSCOPY

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The etching and annealing behaviour of heavy ion nuclear tracks have been studied in glass detectors namely sodalime, phosphate and quartz using optical absorption spectroscopy. All these glass detectors were exposed at 90° and 45° for different ions, with collimated beams of varying energies. The absorption difference spectrum was employed to describe the annealing kinetics. A mathematical relation proposed in our laboratory was used to explain the annealing behaviour of radiation damage due to heavy ion beams in glass detectors. The activation energy was obtained from a new formulation and is found to be 0.16 eV in sodalime, 0.56 eV for phosphate and 0.69 eV for quartz glass detectors.

When glass is exposed to radiation, numerous changes in the physical properties can take place, e.g. optical absorption bands may be introduced, nuclear tracks can be formed, disruption of glass structure can take place, the density of glass can change, paramagnetic defect centres can be formed and the physical strength of a glass can be altered.

It is well known that the irradiation of a material by particles produces not only atomic displacements but also ionizing events. However, Frieble and Griscom[1] maintained that irradiation of glass produces defect centres and explained how these centres are formed on atomic level. They described the two principal interactions of radiations with glass as the ionization of electrons and direct displacement of atoms by elastic scattering. When the glass is subjected to ionizing radiations (α or γ ray photons, ultraviolet light or charged particles), electrons are ionized from the valence band if the energy of the radiation is greater than the band gap and the excess energy is converted to kinetic energy. Then the electrons move through the glass matrix and will either be trapped by pre-existing flaws to form defect centres in the glass structure, recombine with the positively charged holes, or in the case of high energy electrons produce a secondary electron cascade by knock-on collisions with the bound electrons.

The electronic structure of many of the radiation induced defect centres in both glass and crystalline materials is amenable to study by experimental tools such as optical absorption spectroscopy[2]. In fact, the visible absorption often induced by radiation in these materials has led to the term *colour centres* to describe the defect centres.

The irradiation of glass also results in induced optical absorption which can cause the glass to colour. The absorption arises because the splitting between the ground and excited states of the radiation induced defect centres is such that optical phonons iduce transitions between the states.

The important point about radiation effects in glass is that the radiation induced defects studied by various techniques arise because of hole or electron trapping so, there is a probability of thermal untrapping that may be described as:

$$P = S \exp(-E/kT) \tag{1}$$

where S is the escape frequency and E is the height of the barrier[3]. This untrapping can result either in recombination or in retrapping at the same or different defect sites.

Optical Absorption in Glass Detectors

The optical absorption is observed in irradiated glass because incident photon of light have the proper energy to induce a transition line between the ground state and an excited state of the defect center. In theory, the absorption of defect center should be extremely narrow and well defined, but in practice thermal fluctuations and site to site variations result in a distribution of energy level splitting. Thus, the measured spectrum is found to consist of broad lines.

Recently, it has been observed by various authors[4-9] that nuclear tracks obtained due to radiation damage in glasses and minerals show enhanced optical absorption near the intrinsic absorption edges and it was found that this technique is non-destructive which gives additional information about the structure and other properties of radiation induced defects. The position of the absorption edges is slightly dependent on the concentration of defects. Ritter and Mark[9-10] recently observed enhanced optical absorption due to natural radiation damage in apatite. They concluded that the radiation damage must be the source of Characteristic Absorption Difference (CAD). The term CAD from absorption difference spectrum has been used[11-12] because it indicates the difference of the absorption co-efficient of two samples obtained from the logarithmic ratio of the intensities ($\log I/I_o$) transmitted by each of the two samples. The intensity of the absorption is, of course, proportional to the defect center concentration in the glass.

According to Ritter and Mark[13] and Mark and Ritter[14], the increased rate of CAD might be due to the fact that the CAD production is no longer controlled by the increase of atomic displacement but by the carrier production or annihilation at other atomic defects. Such a process could also account for the difference in CAD response of radiation damaged samples.

Thus, in the present study, optical absorption spectroscopy is used for the investigation of the annealing behaviour of heavy ion radiation damage in glass detectors. The experiment is performed in a dual-beam spectrophotometer where, at a given wavelength, both the intensity of light passing through the sample, I, and that of reference beam, I_o, are simultaneously measured. The reference chamber can either be empty or contain an unirradiated sample. The spectropho-tometer, then, ratios these intensities and presents the data as percent transmission: $T = 100 \, I/I_o$. The behaviour of CAD at about 314 nm, between unirradiated and irradiated samples of sodalime glass, has been used for calculating the activation energy of annealing, as a function of annealing temperature at constant time.

The phenomenon that absorbing defects have different annealing characteristics than the etchable heavy ion track defects can be put to use to develop an independent relation for activation energy.

According to Mark and Ritter[14], the peak height obtained by radiation damage in sodium silica glass is proportional to defect concentration and therefore, the peak height reduction during isothermal annealing studies is a measure of the point defect annealing behaviour in a better-fitted single exponential function as

$$A(t) = a \exp(-\alpha t) \tag{2}$$

where A(t) is the absorption after annealing time, t, with an annealing coefficient as

$$\alpha(T) = \alpha_o \exp(-E/kT) \tag{3}$$

and α_o is a pre-exponential coefficient, E, the activation energy, T, the annealing temperature and k, Boltzmann's constant.

However, it has been a subjective matter to find a simpler relation which gives better results. Therefore, in order to calculate the activation energy directly from optical absorption, a well resolved and well understood UV spectra can be utilized by measuring the absorption difference of characteristic spectral features. Hence, we attempted to modify our annealing formulation[15] by replacing annealing rate with a CAD reduction ratio, i.e.

$$(CAD)_r = At^{-n} \exp(-E/kT) \tag{4}$$

where $CAD_r = CAD_i/CAD_f$ with r denoting reduction ratio and the subscripts i, f refer to values before and after annealing temperature, T. A is a proportionality constant, k, Boltzmann's constant, n, the exponent of annealing time t and E is the activation energy of annealing.

The above relation (4) has been tested for annealing experiments in sodalime, phosphate and quartz glass detectors irradiated with heavy ions.

Experimental

The samples of sodalime (microscopic glass slide), phosphate and quartz glass detectors of exactly equal thickness were subjected to heavy ion irradiation with penetrating beams of ^{208}Pb (17 and 13.6 Mev/u) and ^{139}La (14.6 MeV/u) at GSI heavy ion accelerator, UNILAC (GERMANY). The irradiated slides were then cut into equal sizes of 4 cm x 1 cm to match the dimensions of UV- absorption spectrophotometer cell.

Irradiated glass samples were annealed in a Muffle furnace at 200, 250, 300, 350 and 400°C respectively, for time intervals of 15 min, each. The unirradiated sample was taken as a comparison blank in the reference chamber. The absorption spectra of all the thermally-annealed samples was obtained in the wavelength range of 400-200nm. It has been found that at about 314 nm there appears a maximum in the difference between an irradiated and comparison blank (untreated sample). This difference is termed as the Characteristic Absorption Difference (CAD) and is measured from the peak of the curve obtained as shown in (fig. 1). It is observed that CAD decreases with the increase of annealing temperature. Thus the healing of defects during heating is the main cause of CAD reduction.

Discussion of Results

The characteristic absorption difference is found to decrease with the extent of annealing. The CAD for untreated and annealed samples of sodalime glass is shown in Fig. 1 at different annealing temperatures for ^{208}Pb (17 MeV/u) ion beam and the values are summarized in Table 1.

Table 1 - The Characteristic Absorption Difference (CAD) for different ion beams in sodalime glass detector.

Sr. No.	T (°K)	$\frac{10^3}{T(°K)}$	^{208}Pb (17 MeV/u)		^{208}Pb (13.6 MeV/u)		^{139}La (14.6 MeV/u)	
			CAD	CAD$_r$	CAD	CAD$_r$	CAD	CAD$_r$
1	Untreated		69.40		62.40		62.40	
2	473	2.11	49.50	0.71	40.70	0.65	42.00	0.67
3	523	1.91	33.40	0.48	29.80	0.48	31.30	0.47
4	573	1.75	24.40	0.35	21.80	0.34	23.60	0.37
5	623	1.61	18.70	0.27	16.70	0.27	17.60	0.28
6	673	1.49	15.30	0.22	12.50	0.21	12.70	0.22

The above relation (4) is utilized for calculating the activation energy from CAD reduction in sodalime glass. Rewriting the above relation (4), we have

$$\ln CAD_t = \ln A - n \ln t - E/kT \qquad (5)$$

The slopes of the plots of the left hand side of equation (5) against inverse temperature for different ion beams (Figs. 2-4) will yield the activation energy of annealing. Similar plots were obtained for phosphate and quartz glass detectors. The values of activation energies for different ion beams are found to be 0.16 eV in sodalime, 0.56 eV in phosphate and 0.69 eV in quartz glass detectors (Table 2).
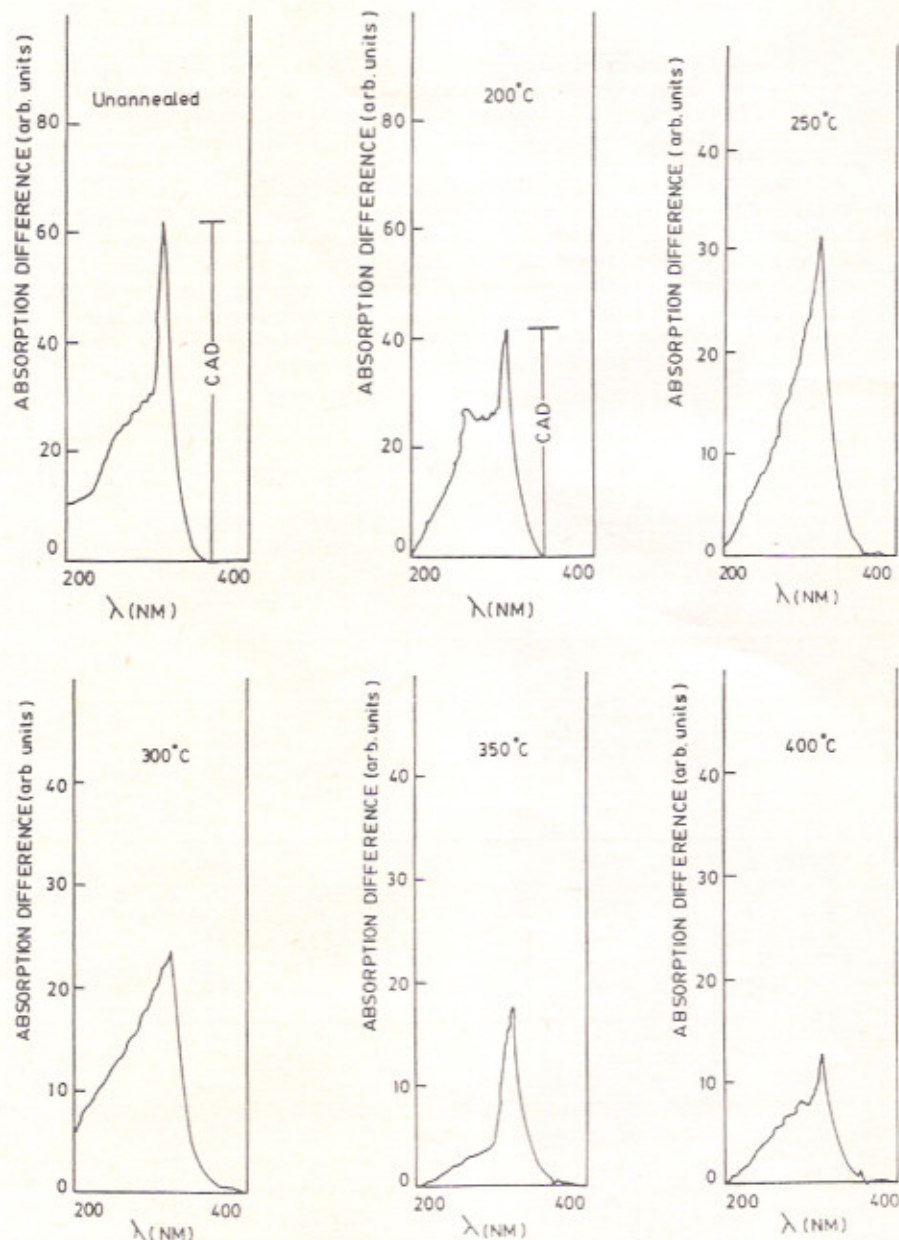
Fig. 1. The Characteristic Absorption Difference (CAD) for
unannealed and annealed samples of sodalime glass
irradiated with ^{139}La (14.6 MeV /n).

Table 2 - The comparison of activation energies in different glasses irradiated with varying ion beams.

Ion Beams	Activation Energy (eV)		
	Sodalime	Phosphate	Quartz
^{208}Pb (17 MeV/u)	0.16	0.56	0.69
^{208}Pb (13.6 MeV/u)	0.155	0.56	0.69
^{139}La (14.6 MeV/u)	0.16	0.56	0.69
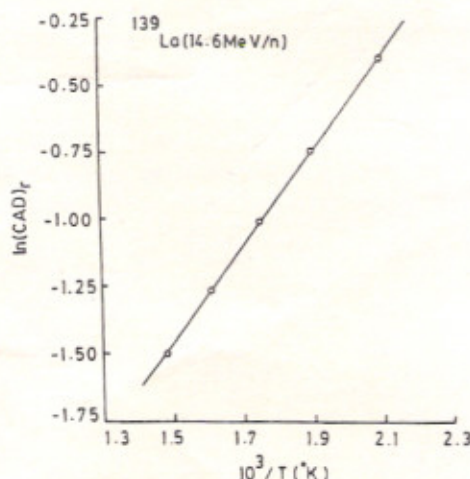
Fig. 2. The plot of ln (CAD), against inverse temperature using ^{139}La (14.6 MeV / n) ion beam in sodalime glass detector
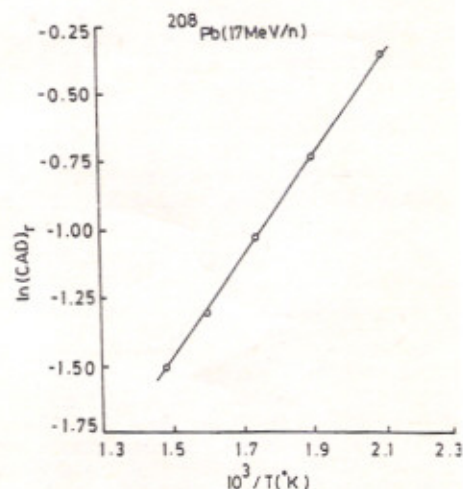
Fig. 3. The plot of ln (CAD), against inverse temperature using ^{208}Pb (17 MeV / n) ion beam in sodalime glass detector.
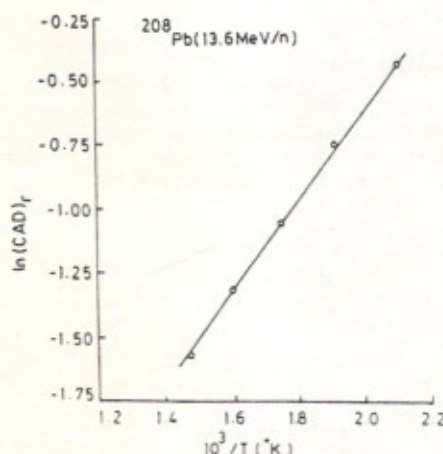
Fig. 4. The plot of ln (CAD), against inverse temperature using ^{208}Pb(13.6 MeV / n) ion beam in sodalime glass detector.

Conclusions

Therefore, we may conclude that:

1. Identical values of activation energies obtained for different radiation sources implies that similar elementary process are operative for the annealing of radiation damage caused by the different ionizing radiations.

2. The new approach of calculating the activation energy using absorption spectroscopy further strengthens the concept that a single activated process is involved during annealing of a radiation damaged glass.

3. The present results are in agreement with our previous findings[16,17].

4. While constructing empirical formulations, the simple formulations with less fitted parameters are generally preferable.

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