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Solid State Nuclear Track Detectors in the Measurement of Alpha to Fission Branching Ratios of Heavy Actinides

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Abstract

A sequential etching procedure for revelation of alpha and fission tracks in CR-39 was developed and optimized . Using this technique alpha and fission tracks can be differentiated unambiguously because of significant differences in their sizes and etching times . This registration and revelation procedure for alpha and fission tracks may be used for the studies of half lives , alpha to fission branching ratios and identification of radionuclides based on their decay schemes . It has the added advantage that both alpha decay and fission events can be studied using one detector and hence uncertainties related to efficiency ,registration geometry, registration times, amount of radionuclides etc can be eliminated or minimised . The effects of neutron , gamma and alpha radiations on the alpha and fission fragment tracks registration and revelation properties of CR-39 detectors [CR-39,CR-39 (DOP)] were also studied . The IR spectra were also studied to find out the nature of chemical changes produced by these radiations on CR-39 . The sequential etching procedure was used to measure the α/S.F. branching ratio of ^{252}Cf and ^{244}Cm and compared with published data in which other techniques were used . These two radionuclides were chosen because in ^{252}Cf both alpha and spontaneous fission rates are

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comparable and in ^{244}Cm alpha events are much more than spontaneous fission events and hence form two extremes . The results indicate that the present method gives very precise and accurate values for α /S.F. branching ratio for ^{252}Cf and ^{244}Cm and can be considered as reference nuclides for this technique of measurement.

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Introduction

Solid state nuclear track detectors (SSNTDs) have been used very effectively in the accurate measurement of both extremely large and extremely short spontaneous fission half lives. When one considers spontaneous fission decay of heavy elements it is important to realize that alpha decay is a competitive process. Consequently, the heavy element isotopes are associated with intense alpha activity causing problems in handling and those arising from the pile-up alpha pulses. For example, the ratio of alpha to spontaneous fission decay rate of ^{244}Cm is about 7.1×10^5. The determination of long SF life-times require fairly large amounts of the isotopes and large measurement times. The vast amount of data on decay schemes of radionuclides have been generated by various researchers using different techniques and methods. But the present situation regarding our knowledge of half lives of some heavy element isotopes is somewhat confusing because of discrepancies in the data. One example is the total half life of ^{252}Cf. The available information on this important datum has been summarized by Smith[1] , who has also provided an informative analysis of potential problems associated with many of the individual measurements. The available data appear to cluster around two distinct values for the ^{252}Cf half-life viz.

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2.638 yr. and 2.651 yr. Since evidence exists which provides support for the correctness of each of these values, it is not clear which of them represents the true ^{252}Cf half life. However, such a situation can be clarified by accurately measuring the alpha emission to spontaneous fission branching ratio along with one of the partial half lives.

Most of the measurements of α/F branching ratio of Californium and Curium isotopes have been carried out using semi conductor detectors (pulse height analysis)[2-6]. MacMurdo[6] used lexan plastic detector and 2π fission chamber method for fission fragment detection and surface barrier detector for alpha emission measurements. Aleksandrov[4] measured the α/F branching ratio of ^{252}Cf using thick layer photo emulsion detector. In the measurements of α/F branching ratio using semi-conductor detector, fission fragment pulses are separated from alpha pulses by pulse height analysis. In this experiment, most of the error comes from the uncertainty in interpretation of those parts of the spectra which could not be unambiguously attributed either to α-particles or to fission fragments. Moreover, if α/F ratio is high, then spontaneous fission events should be measured with another technique to avoid statistical errors. In such situations the integrating nature of SSNTD coupled with their insensitivity to alpha particles (lexan and mica in particular) are major advantages over the conventional electronic techniques. If separate detectors are used for measurements of alpha and spontaneous fission fragments, then there is a possibility of introducing more errors because of difference in the efficiencies of two different techniques, change in particle registration geometry etc. Using same detector for both alpha and fission fragments registration will be advantageous because many measurement factors and conditions will be similar for both alpha and fission fragments. Aleksandrov[4] used thick layer photo emulsion detector which can integrate events for long periods. But low sensitive detector has to be used for easy identification of α-particles and fission fragments. Moreover, the particles which enter the emulsion at a right angle or at angles close to a right angle are generally difficult to identify. The error may also be introduced in results of analyzing the spectra of the mean free paths of the fission fragments and α-particles in photo-layer. Photo-emulsion detectors are now replaced by more sensitive and versatile solid state nuclear track detectors like lexan, CR-39, CN, mica etc.

Charged particles ranging from proton to fission fragments can be registered with high sensitivity in CR-39 solid state nuclear track detectors. We have used this unique property of CR-39 along with sequential development of alpha and fission tracks for the measurements of α/F branching ratios of radionuclides, namely ^{252}Cf and ^{244}Cm. Using a sequential etching procedure, it is possible unambiguously to reveal both alpha particles and fission fragments. This method is highly sensitive and simple as it does not involve complicated electronic circuitry for the measurements of α/F branching ratio. This procedure enables one to measure both alpha particles and fission fragments with the same detector and hence it eliminates the uncertainties related to geometry, efficiency and exposure time. Since this method is also amenable for 'on line' use, the radionuclides produced in submicrogram quantities having very short life times can be studied and identified based on their α and spontaneous fission decay schemes. This detection procedure of α and fission fragments can be used in experimental arrangements like 'moving belt method' [Flerov et.al[7]] or 'rotating drum method' [Hulet et.al[8]] for the studies of trans-fermium isotopes with the added advantage that both alpha and fission can be registered and unambiguously separated from each other.

In this report, we present the details of optimization of the method along with results of systematic studies on the effects of extraneous radiations on the alpha and fission fragments track registration and revelation properties of CR-39. The results of α/S.F. branching ratio measurements of ^{252}Cf, ^{244}Cm are also discussed in this report.

Experimental

1. Optimization of the method

For optimizing the sequential etching procedure, the CR-39 (DOP) solid state nuclear track detector pieces of size 2 cm^2 were exposed to a 1 nanogram planchetted ^{252}Cf source at 2π geometry. These detectors were etched in 6N NaOH at 70°C and 60°C temperature with constant stirring using a magnetic stirrer. These detectors were scanned at 15 min. etching time intervals upto 2 hrs. and after that at 30 min. etching time intervals upto 5 hrs.under an optical microscope at 100 x magnification to count the etched tracks.The average track diameters were also measured under optical microscope. At each etching temperature, the graph between etching time vs. track density was plotted. Lexan detectors were also exposed to ^{252}Cf at 2π geometry and were etched under the same etching condition (6N NaOH, 60^0 & 70^0C temp.) at definite etching time intervals. The lexan detector is insensitive to alpha particles and hence it provided the fission fragment track densities.

2. Effects of radiation on the registration efficiency of CR-39

2.1 Neutron effects

In order to study the effects of neutron fluence on CR-39, the bulk etch rate (V_G) of neutron irradiated and un-irradiated CR-39 detectors were measured. Irradiation of CR-39 detectors were done at the APSARA swimming pool reactor. Two types of detectors, CR-39 and CR-39 (DOP) were irradiated from 2 minutes to five hours. The neutron fluxes were experimentally determined by using the method developed by Iyer et. al[9] . In this method, fission tracks were recorded in small strips of lexan detectors immersed in a uranium standard solution which was irradiated with neutrons. CR-39 detectors were irradiated in the same irradiation environment simultaneously. The neutron flux was calculated from the following equation:

$$Td = \frac{K_{wet}\ C.N.\ \sigma\ \phi\ t}{A} \qquad \text{--------(1)}$$

where T_d is track density (the number of tracks per Cm^2); K_{wet} , 'track registration efficiency' in solution (8.1×10^{-4} cm for lexan); 'C' the concentration of solution uranium standard

(g/cm^3); A the atomic weight of uranium ; σ, the fission cross section (cm^2); ϕ, the neutron flux ($\#/cm^2/sec$) and t, time of exposure (sec) to neutron flux.

All the CR-39 detectors were irradiated in a reactor at a fixed position so as to keep all the conditions similar for every CR-39 detector. Etching of CR-39 (both irradiated and unirradiated) was done at 70°C in 6N NaOH and for 1 hr. with constant stirring using a magnetic stirrer. All the detectors were etched simultaneously so as to keep similar conditions for every detector. After and before etching of CR-39 detectors, the thickness, weights, perimeter and area of CR-39 detectors were measured. The bulk etch rates V_G ($\mu m/hr.$) of CR-39 detectors were calculated from the following equation[10].

$$V_G = \frac{(m_1-m_2)}{2m_2} \quad t_2 \quad \left[1 - \frac{Pt_2}{2A} \right] \qquad \text{-------(2)}$$

where m_1 is detector mass before etching, m_2 mass after etching, t_2 the detector thickness after etching, P the detector perimeter and A the detector area.

The infrared transmittance spectra of irradiated detector sheets and unirradiated sheets were taken using a Perkin-Elmer 577 infrared spectrometer. All the spectra (IR) were recorded with attenuator, as the original transmittance was quite low. The CR-39 detectors were irradiated with neutron flux of the order of 7.46×10^{10} $/cm^2/sec$ in APSARA from 10 minutes to 5 hrs. The thickness of unirradiated and irradiated detectors were same, 0.5 mm. The percentage transmittance of CR-39 detectors were converted into absorbance and plotted against irradiation time.

For the study of variation of track densities with neutron flux, the CR-39 (DOP) detectors were irradiated with neutrons at known fluxes in APSARA reactor at a fixed distance from the core of the reactor for periods ranging from 2 min. to 2 hours. After neutron irradiation, the detectors were exposed to ^{252}Cf in 2π geometry for 2 hrs. to register the alpha and fission tracks. The irradiated and unirradiated detectors were etched in 6N NaOH, 70°C for 30, 60, 120, 180 min. etching time to reveal alpha and fission tracks. The track densities were evaluated by counting the tracks in the CR-39 detectors under an optical microscope at 100 x magnification. To find out the effects of fast neutrons and thermal neutrons, the CR-39 (DOP) detector sheets were also exposed to thermal neutrons at the thermal column in APSARA reactor. The irradiated and unirradiated CR-39 (DOP) detectors were exposed to ^{252}Cf in 2π geometry to register the alpha and fission tracks. The tracks were revealed by etching the detectors in 6N NaOH at 70°C and using a magnetic stirrer. The fission tracks were counted under an optical microscope after 1 hr. etching and α-tracks were counted after 3 hrs. etching. The thermal neutron flux was measured by using the method described above (Iyer et.al[9]) and found to be 4.1×10^{6} /cm^{2}/sec. The CR-39 detectors were exposed to thermal neutrons for 12 hrs. to 48 hrs.

2.2 Gamma Radiation effects on CR-39

The CR-39 (DOP) pieces each of size 2 cm^{2} were cut from a big sheet. These samples were irradiated with ^{60}Co gamma rays in the gamma irradiator in the Radiochemistry Division, CARC for different doses of 5.5 M Rad , 17.9 M Rad , 40.5 M Rad and 53.0 M Rad. The detectors were etched in 6N NaOH, 70°C for 1 hr. and bulk etch rates of unirradiated and irradiated CR-39 (DOP) detectors were measured as described above. The pre-irradiated CR-39 (DOP) detectors were then exposed to ^{252}Cf source along with the unirradiated detector in 2π geometry. These detectors were then etched for different intervals of time in 6N NaOH at 70°C to reveal alpha and fission tracks. The track densities were measured by counting the tracks under an optical microscope.

2.3 α-Radiation effects on CR-39

In order to study the effects of alpha radiation on fission track registration and revelation efficiency of CR-39 (DOP), the CR-39 (DOP) detector pieces of 2 cm^2 area were exposed to an alpha source of ^{244}Cm in 2π geometry from 1 hr. to 14 days. The alpha flux of ^{244}Cm in 2π geometry was determined experimentally using solid state nuclear track detectors like cellulose nitrate, CR-39 and alpha proportional counters. The alpha flux of ^{244}Cm in 2π geometry was found to be 3.1×10^5 alphas/cm^2/min. The cellulose nitrate and CR-39 detectors record fission events also which occur along with alpha decay in ^{244}Cm. In such cases alpha track densities were corrected by substracting fission track densities which in turn were obtained by fission-specific and more radiation resistant mica and lexan track detectors. The alpha pre-irradiated CR-39 (DOP) detector pieces were exposed to ^{252}Cf for 2 hrs. These exposed detectors were etched in 6N NaOH, 70°C and for different time intervals upto 1 hr. The fission track densities of exposed CR-39 (DOP) detectors were measured by counting under an optical microscope. The track densities of α-exposed detectors were compared with unirradiated CR-39 (DOP) detector which was exposed to ^{252}Cf and etched under similar conditions.

3. Measurements of α/S.F. branching ratio of ^{252}Cf and ^{244}Cm

3.1 α/S.F. branching ratio of ^{252}Cf

The two types of CR-39 detectors CR-39 and CR-39 (DOP) were exposed to an electrodeposited source of ^{252}Cf approximately 2 X 10^{-13} g/cm^2 thickness in 2π geometry. The detectors were kept 6.5 mm above the ^{252}Cf source in air to avoid contamination of the detectors and also to avoid particles entering extreme low angles in the detector (below 10° angle) which may be below the critical angle of CR-39 or the angle in which particles during traveling in the source itself may lose their energies and may not produce etchable tracks. The detectors were exposed to ^{252}Cf source from 1 hr. to 4 hrs. The CR-39 (DOP) detectors were etched in 6N NaOH at 70°C for 30 min. and 120 min. to reveal fission and alpha tracks respectively. These were counted under an optical microscope to get fission and alpha track densities. The CR-39 detectors were etched in 6N NaOH at 70°C for 45 min. and 210 min. to reveal fission and alpha track respectively. The background tracks (mostly alpha tracks) were measured by etching blank detectors under similar conditions and the background track densities were subtracted from fission and alpha track densities.

3.2 α/F branching ratio of ^{244}Cm

The CR-39 and CR-39 (DOP) detector pieces of 2 cm^2 area were exposed to a 3.5 ngm/cm^2 planchetted ^{244}Cm source in 2π geometry. The exposure time varied between 30 sec. to 1 min. for measuring α track densities while for fission track densities it varied from 172 hrs. to 330 hrs. The distance between detector and ^{244}Cm was kept at 6 mm. The etching and measurements of track densities were carried out as described above. The CR-39 detectors were etched for 45 min. and 4 hrs. to reveal fission and alpha tracks respectively. The CR-39 (DOP) detector were etched for 30 min. and 4 hrs. to reveal fission and alpha tracks respectively.

4. Results and discussion

4.1 Development of sequential etching procedure for alpha and fission tracks in CR-39

CR-39, diethylene glycol bis allyl carbonate, is a very homogeneous polymer and it can register a range of charged particles. Ever since the discovery of the track recording properties of CR-39[11] , a number of papers have been published about its etching characteristics, registration and response to protons, light and heavy charge particles[12,13,14]. The tracks of different charged particles are formed at different depths in CR-39 depending upon their mass, charge, energy or energy loss rate (dE/dx) in the detector. The charged particles can be identified in solid state nuclear track detectors based on the track parameters. But for measurements like half lives or α/S.F.

branching ratio, one has to accumulate a large number of tracks to reduce statistical uncertainties. In such cases the identification of each and every track may not be practically possible. However, this problem can be avoided if there is <u>significant difference</u> in the sizes of tracks of alpha and fission fragments or with appearance times of tracks on etching of detectors (difference in etching time). Cellulose nitrate and CR-39 detectors can register both alpha particles and fission fragments. To study the possibilities of revealing alpha and fission fragment tracks in cellulose nitrate unambiguously, the CN detectors were exposed to ^{252}Cf in 2π geometry for 2 hrs. to register alpha and fission fragments. Both alpha and fission tracks were revealed after etching in 6N NaOH at 60°C for 40 min. It was found that alpha tracks were small and circular shaped and have 2.6 μm to 6.5 μm diameter and fission tracks were found to be having 6.5 μm to 13 μm length , conical and needle shaped . This indicates that there is not much difference in sizes of alpha and fission tracks and their appearance time in detectors. This fact became more clear when the tracks were counted. It was found that the fission tracks densities in cellulose nitrate detectors were found to be more than those in lexan detectors which records only fission tracks and track densities in different CN detectors were not consistent. The α/F branching ratio were found to be varying from 14.8 to 12.8 which was expected to be about 15.75 in 2π geometry for ^{252}Cf. Obviously some of the alpha tracks might have been counted as fission tracks because there is no significant difference in sizes of alpha and fission tracks. By changing the concentration of etchant or temp., this situation could not be improved.

CR-39 was found to be better than cellulose nitrate detectors for unambiguous separation of alpha and fission tracks because of significant differences in their sizes and etching times (appearance time on etching the detector). The fission tracks appear in CR-39 (DOP) and CR-39 after etching the detector in 6N NaOH, 70°C for 35 min. and 45 min. respectively. The alpha tracks in CR-39 (DOP) and CR-39 appear after etching for 100 min. and 150 min. respectively. The etching time of alpha and fission tracks at different temp. is given in Fig.1. From Fig.1 it is clear that there is a significant difference in etching time of fission tracks and alpha tracks. Hence, fission tracks can be counted first and then after more etching, total track density (α and fission track density) can be measured. Additionally there is significant difference in the sizes of alpha and fission track also. For example, when the CR-39 (DOP) and CR-39 detectors exposed to ^{252}Cf in 2π geometry at 6.5 mm distance between source and detector, were etched in 6N NaOH, 70°C and for 210 min., the fission tracks and alpha tracks diameters were found to vary from 7.8 μm to 13 μm (Av. 10.4 μm) and 6.5 μm to 13 μm (Av. 7.8 μm) for fission tracks and 2.6 to 5.2 μm (Av. 3.9) and 2.6 to 3.9 μm (Av. 3.2 μm) for alpha tracks respectively in CR-39 (DOP) and CR-39 detectors. Photomicrograph (Fig.2) of alpha and fission tracks also makes it clear that alpha and fission tracks can be discriminated easily in CR-39 detectors. Therefore, alpha and fission track densities can be measured unambiguously by counting both the tracks simultaneously.

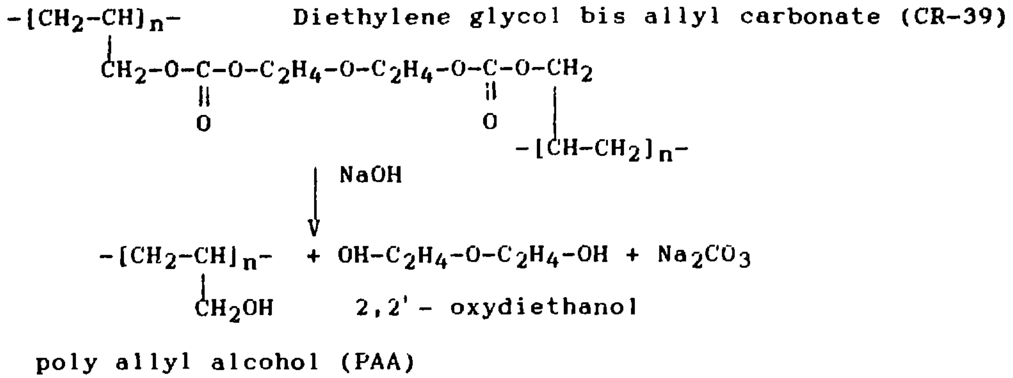
This procedure was optimized by carrying out etching at different etchant conc., temp. and time. It was found that 6N NaOH, 70°C temp. and 180 to 210 min. etching time were the best conditions for developing alpha and fission tracks because it involves less etching time and ensured effective discrimination of α and fission tracks having different sizes. This method of measurements of α and fission tracks may be useful for the studies of half lives, α/F branching ratio, identification of radionuclides based on their decay schemes because with one detector both α and fission events can be measured and hence uncertainties related to efficiency, registration geometry, registration time, amount of radionuclides etc. can be eliminated/minimized.

4.2 Effects of extraneous radiations on physical and chemical properties of CR-39 detectors

In many accelerator/nuclear reactor-based experiments, the detectors are exposed to large doses of neutron, beta particles and gamma rays and the effect of these extraneous radiation on the track registration and revelation properties of CR-39 is important for wider application of this sequential etching procedure. The studies of Blatchley et.al[16] showed that at doses as low as 1 M Rad, the development of tracks from alpha particles was markedly affected by the gamma irradiation background. The average track diameter of alpha tracks in CR-39 was increased from 4.43 ±0.015 μm in unirradiated detector to 8.67 ±0.10 μm in irradiated CR-39 with a gamma dose of 1.2 M Rad. An immediate consequence of this result is that exposures made near/in an accelerator must be kept short if particle identification on the basis of range and linear energy transfer, as reflected in etched track diameter, is to be made possible. Moreover, some of the alpha tracks in high irradiation background may etch to such large diameters that they masquerade as heavier fission fragments and hence may cause problems in counting the fission fragments. However, such a situation can be avoided if effects of extraneous radiations on registration and revelation properties of CR-39 are known.

The effects of gamma irradiation on bulk etch rate of CR-39 are well studied[16-24] mostly with the central objective of using it as a dosimeter. From these studies, it is clear that τ-irradiation enhances the bulk etch rate of CR-39 and bulk etch rate is dose-rate dependent. The studies on effects of electron irradiation[20,25] and neutron irradiation[19,26,27] on bulk etch rate of CR-39 also indicate that electron and neutron irradiation also enhance the bulk etch rate of CR-39. We have also found that τ-irradiation and reactor neutron irradiation enhance the bulk etch rates of CR-39 and CR-39 (DOP) and the ratio of bulk etch rate of irradiated to unirradiated detector is linearly dependent on dose (see Fig. 3a and 3b).

When radiations interact with CR-39, they produce some damage in the detector. The damages caused by the radiations could be due to the production of radicals . The polymer chain is broken (chain scission). It was found by Stenjny et.al.[28] that the length of poly allyl chain was not changed in ^{60}Co τ-irradiation in CR-39 plastic while the etch rate significantly increased. This shows that the poly allyl chains are relatively inert and not responsible for the radiation sensitivity of CR-39 and that it is the diethylene glycol dicarbonate links which are damaged by radiation. The radical produced in the CR-39 from radiation interact with O_2 diffusing from air[29] and cause linear dependence of bulk etch rate on dose[18]. The linear dependence of bulk etch rate on irradiation dose may also be explained in terms of a radiation induced growth in the surface concentration of chain ends[20]. The CR-39 detectors [CR-39 & CR-39(DOP)] were found to acquire an yellow tinge on low neutron flux irradiation and brownish colour on high neutron flux irradiation. These colour changes may be due to electrons, excited by the radiations to essentially free states in the conduction band, especially at trapping sites[30]. The infrared transmittance spectra of the reactor-neutron-irradiated CR-39 detectors were compared with that of unirradiated CR-39 detectors. Only a few bands of frequencies were transmitted to a considerable extent by both irradiated as well as unirradiated CR-39 detectors and peaks were found to be at the same position in both unirradiated and reactor neutrons irradiated CR-39 (see Fig.4). Apparently, there is no change in chemical structure of the reactor neutron irradiated detectors. But the change in absorbance of neutron irradiated CR-39 were found in two peaks at 3200 cm^{-1}, 660 cm^{-1} and in a valley at 2340 cm^{-1}. A linear dependence of absorbance on neutron flux was observed at all these wavenumbers (see Fig.5) . This may be due to breaking of carbonate inter linkage which shorten the chain length and hence increases the end chain concentration. This is also corroborated by the linear dependency of bulk etch rate on neutron flux (Fig. 3a). Similar mechanism was also observed[28,31] in chemical etching of CR-39 by alkalies.



$$\underset{\textstyle CH_2-O-\underset{\textstyle O}{\underset{\|}{C}}-O-C_2H_4-O-C_2H_4-O-\underset{\textstyle O}{\underset{\|}{C}}-O-CH_2-[CH-CH_2]_n-}{-[CH_2-CH]_n-} \xrightarrow{NaOH} \underset{\textstyle CH_2OH}{-[CH_2-CH]_n-} + OH-C_2H_4-O-C_2H_4-OH + Na_2CO_3$$

4.3 Effects of extraneous radiations on the characteristics of alpha and fission tracks in CR-39 detectors

To find out the effects of extraneous radiations on alpha and fission fragment tracks, the variation of fission and alpha track densities in irradiated CR-39 detectors was studied and compared with unirradiated CR-39 detectors. The results are summarized in Table-1. It is obvious from Table-1 that thermal neutron fluence upto 7.01×10^{11} neutrons/cm^2 does not affect the alpha and fission track densities. In case of τ-irradiated CR-39 detector, there is a slight decrease in fission track density at 5.9 M.Rad dose and above this dose , it remains constant. However alpha track densities in τ-rays irradiated CR-39 detectors show a slight decrease of the order of 8 % at a dose of 15.5 M Rad as compared to unirradiated detectors .

The variation of fission track densities in CR-39 and CR-39 (DOP) with integrated reactor neutron fluence of more than

8.95×10^{12} /cm^2 and 1.23×10^{14} /cm^2 respectively are shown in Figure-6. At a neutron fluence about 2.69×10^{14} /cm^2, the CR-39 detectors on etching in 6N NaOH, 70°C became opaque and visibility of fission tracks was very poor. The visibility of fission tracks in reactor neutron irradiated CR-39 could be improved by applying oil on the surface of detectors. However, alpha tracks in neutron irradiated CR-39 at neutron fluence more than 8.95×10^{12} /cm^2, could not be separated from high background. From Fig.6, it appears that fission track densities decrease exponentially with increase in reactor neutron fluence. It was observed that fission track length decreases with increase in neutron flux. For example, in CR-39 exposed to ^{252}Cf at 6.5 mm distance from source in 2π geometry, the track length was found to be 5.2 μm in CR-39 a at reactor neutron fluence of the order of 4.5×10^{13} /cm^2 and 10.4 μm in unirradiated CR-39 detector while all the conditions were kept constant (6N NaOH, 70°C, 30 min.). This indicates that while bulk etch rate increases with neutron influence, the track etch rate is either unaffected or decreases with neutron flux and hence etching response (V_t/V_G), critical angle ($\mathrm{Sin}^{-1}\, V_t/V_G$) and registration efficiency ($1-\sin Q_c$) are adversely affected by radiations. The exponential decrease in fission track densities with increase in neutron flux (Fig.6) indicates that as neutron fluence increases the damage induced by neutron fluence reaches a saturation point. Because of the increase in bulk etch rates with increase in radiation dose, the etching time is also reduced as reflected in Table-1. As shown in Fig.3a, the ratio of V_G(irradiated)/V_G(unirradiated) is more in CR-39 than that of CR-39 (DOP) for same reactor neutron fluence. The decrease in fission track densities with increase in neutron fluence (Fig.6) in CR-39 is faster than CR-39 (DOP). It is obvious from these results that doping with dioctyl phthalate improves the radiation resistance of CR-39 detectors. The study was also made to find out the effects of alpha flux on fission track densities in CR-39. It was observed that at alpha flux of the order of 9.36×10^6 /cm^2, the fission track density is reduced by 11% and thereafter it remained constant (Table-1).

4.4 Application of sequential etching procedure of alpha and fission tracks for measurements of α/SF branching ratio

Finally, this sequential etching procedure was used to measure the α/S.F branching ratio of ^{252}Cf and ^{244}Cm and compared with the published data[3,4,5,15] in which other techniques were used. These two radionuclides were chosen because in ^{252}Cf both α and S.F. events are comparable and in ^{244}Cm α events are much more than spontaneous fission events and hence form two extremes. The data in Tables-2,3,4 show that the present method gives very precise and accurate value for the α/SF branching ratio. The decay products formed in ^{252}Cf, ^{244}Cm do not interfere in measurements of α/S.F branching ratio of ^{252}Cf, ^{244}Cm. Generally, both α and fission fragments are recorded simultaneously from the same source under the same experimental geometry and for the same exposure time. This is possible if the two half lives (for α and SF) are not very widely different from each other as for example in ^{252}Cf. In the case of ^{244}Cm, since the half lives differ from each other by several orders of magnitude, this simple approach was not applicable and needed two independent exposures. For measurement of fission events, the detectors have to be exposed for longer time. Consequently alpha flux would be considerably higher. Another difficulty arises because of the possible non-uniformity of the ^{244}Cm deposit necessitating the scanning of as large an area (preferably the entire exposed area) as possible. With proper understanding of these practical problems and taking the necessary care, it is possible to use the present method to obtain accurate values for this ratio, as clearly indicated by the results of this as well as our earlier studies[32].

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Table-1

Variation of fission and alpha track densities in radiation exposed CR-39 (DOP) detectors

Radiation	Flux	Etching time (min.)	Track density #/cm^2	
			fission	alpha
γ (^{60}Co)	0.00	180	$1.97 \pm .03 \times 10^4$	$3.22 \pm .03 \times 10^5$
	5.93 M Rad	60	$1.70 \pm .03 \times 10^4$	-
		180	$1.71 \pm .02 \times 10^4$	$3.19 \pm .03 \times 10^5$
	15.50 M Rad	60	$1.61 \pm .02 \times 10^4$	-
		120	$1.73 \pm .02 \times 10^4$	$2.95 \pm .03 \times 10^5$
α (^{244}Cm)	0.00	60	$1.53 \pm .03 \times 10^4$	-
	3.15×10^5 /cm^2	60	$1.46 \pm .02 \times 10^4$	-
	9.36×10^6 /cm^2	60	$1.36 \pm .03 \times 10^4$	-
	1.87×10^7 /cm^2	45	$1.36 \pm .04 \times 10^4$	-
	4.49×10^8 /cm^2	45	$1.37 \pm .03 \times 10^4$	-
	6.29×10^9 /cm^2	30	$1.36 \pm .03 \times 10^4$	-
n (thermal neutrons)	0.00	180	$9.37 \pm .09 \times 10^3$	$14.87 \pm .15 \times 10^4$
	1.75×10^{11} /cm^2	180	$9.39 \pm .07 \times 10^3$	$14.84 \pm .11 \times 10^4$
	3.51×10^{11} /cm^2	180	$9.33 \pm .11 \times 10^3$	$14.74 \pm .11 \times 10^4$
	5.26×10^{11} /cm^2	180	$9.36 \pm .05 \times 10^3$	$14.73 \pm .09 \times 10^4$
	7.01×10^{11} /cm^2	180	$9.33 \pm .09 \times 10^3$	$14.77 \pm .13 \times 10^4$

Table-2

α/S.F branching ratio of ^{252}Cf

S.No.	Detector	Fission tracks in specified area	Alpha tracks in specified area	α/S.F in 2π geometry	α/S.F in 4π geometry
1	CR-39(DOP)	1.81×10^4	28.40×10^4	15.69	31.38
2	CR-39(DOP)	1.83×10^4	29.05×10^4	15.87	31.75
3	CR-39(DOP)	1.86×10^4	29.58×10^4	15.90	31.80
4	CR-39(DOP)	2.04×10^4	32.13×10^4	15.75	31.50
5	CR-39	1.67×10^4	26.68×10^4	15.97	31.95
6	CR-39	521	8073	15.50	31.00

Average = 31.56 ± 0.35

Table-3

α/S.F branching ratio of ^{244}Cm

S.No.	Detector	Exposure time	Area counted mm^2	No.of tracks	$\#/cm^2/$ min	α/ SF ratio	
						2π	4π
1	CR-39(DOP)	10305 min	48.92	4513[a]	0.895	-	-
		30 sec.	6.61	10298[b]	3.116×10^5	3.48×10^5	6.98×10^5
		30 sec.	7.43	11620[b]	3.128×10^5	3.495×10^5	6.99×10^5
2	CR-39	19834 min	34.48	5396[a]	0.789	-	-
		1 min.	7.14	19657[b]	2.753×10^5	3.489×10^5	6.98×10^5

a = fission tracks
b = alpha tracks

Av. α/S.F ratio = $(6.977 \pm .0141 \times 10^5)$

Table-4

Comparison of the present method with the other methods

S.No.	Nuclide	Recording method	α/S.F ratio 4π geometry	Ref.
1	^{252}Cf	Semi conductor detector	31.3±0.2	3
2		"	31.5±0.3	4
3		Thick layer photo emulsion	31.1±0.3	4
4		CR-39 (SSNTD)	31.56±0.35	Present Work
5		IAEA value (Based on av. of all published data on half lives of ^{252}Cf	31.32±0.16	15
1	^{244}Cm	Semi conductor method	$7.43\pm.01\text{x}10^{5}$	3
2		Silicon surface barrier detector	$7.420\pm.035\text{x}10^{5}$	5
3		CR-39 (SSNTD)	$6.977\pm.014\text{x}10^{5}$	Present Work
4		IAEA value (based on av. of published data on half lives of ^{244}Cm)	$7.18\pm0.55\text{x}10^{5}$	15

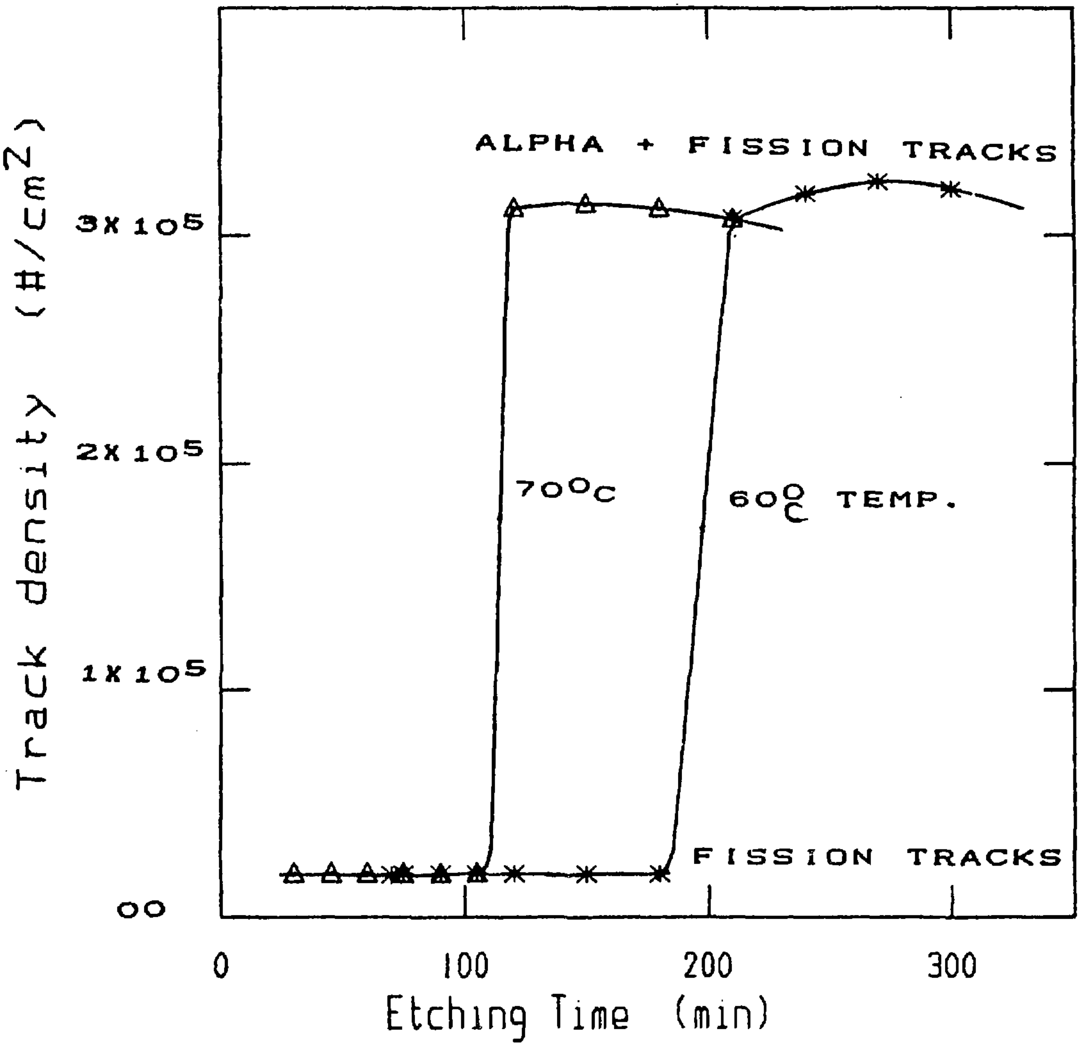

FIG. 1 REVELATION OF ALPHA AND FISSION TRACKS IN CR-39(DOP)

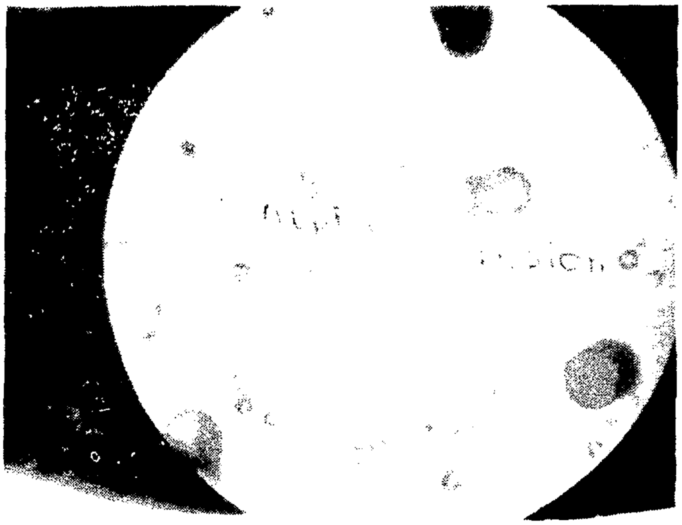

Fig. 2. Photomicrograph of Alpha and Fission Tracks in CR-39

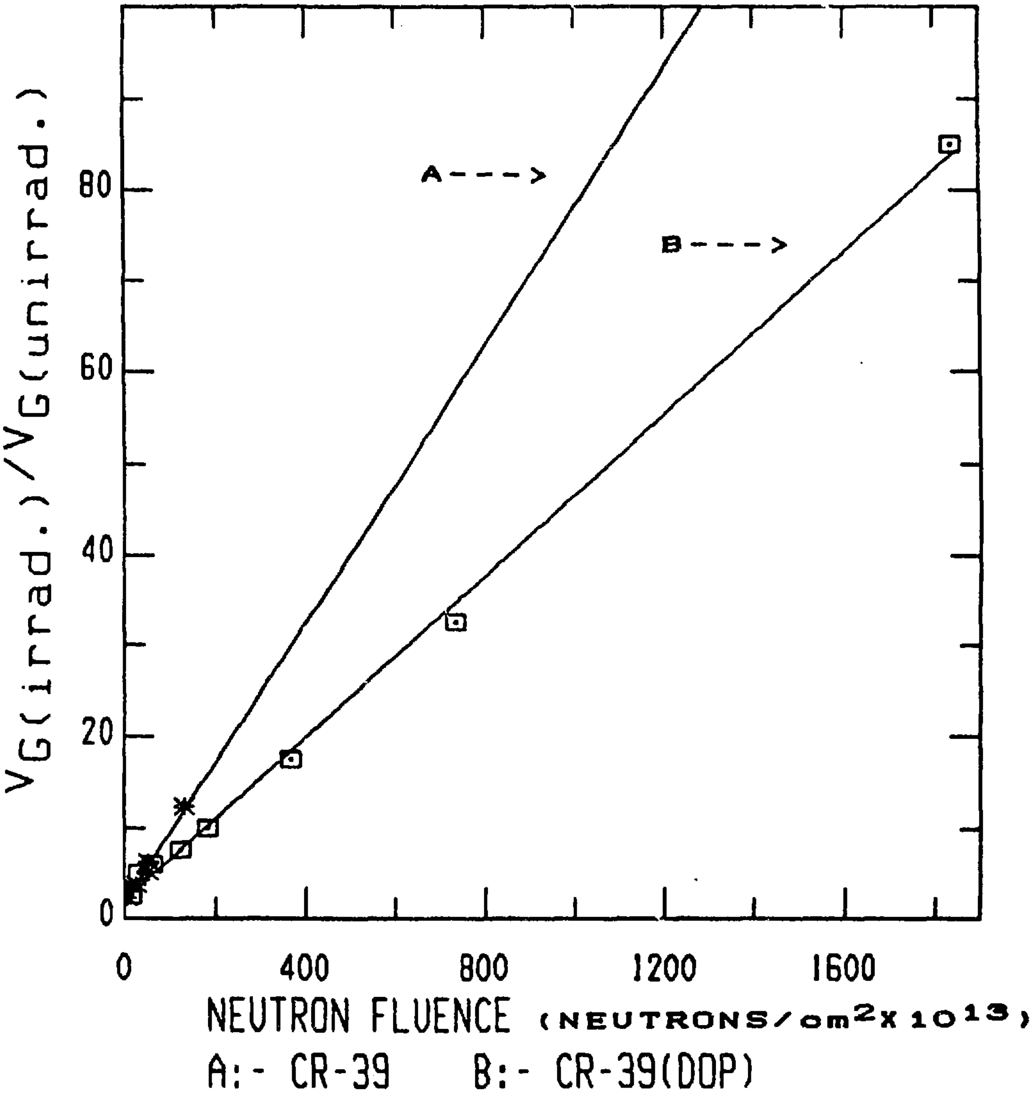

FIG. 3a BULK ETCH RATE OF CR-39 DETECTOR EXPOSED TO REACTOR NEUTRON FLUENCE

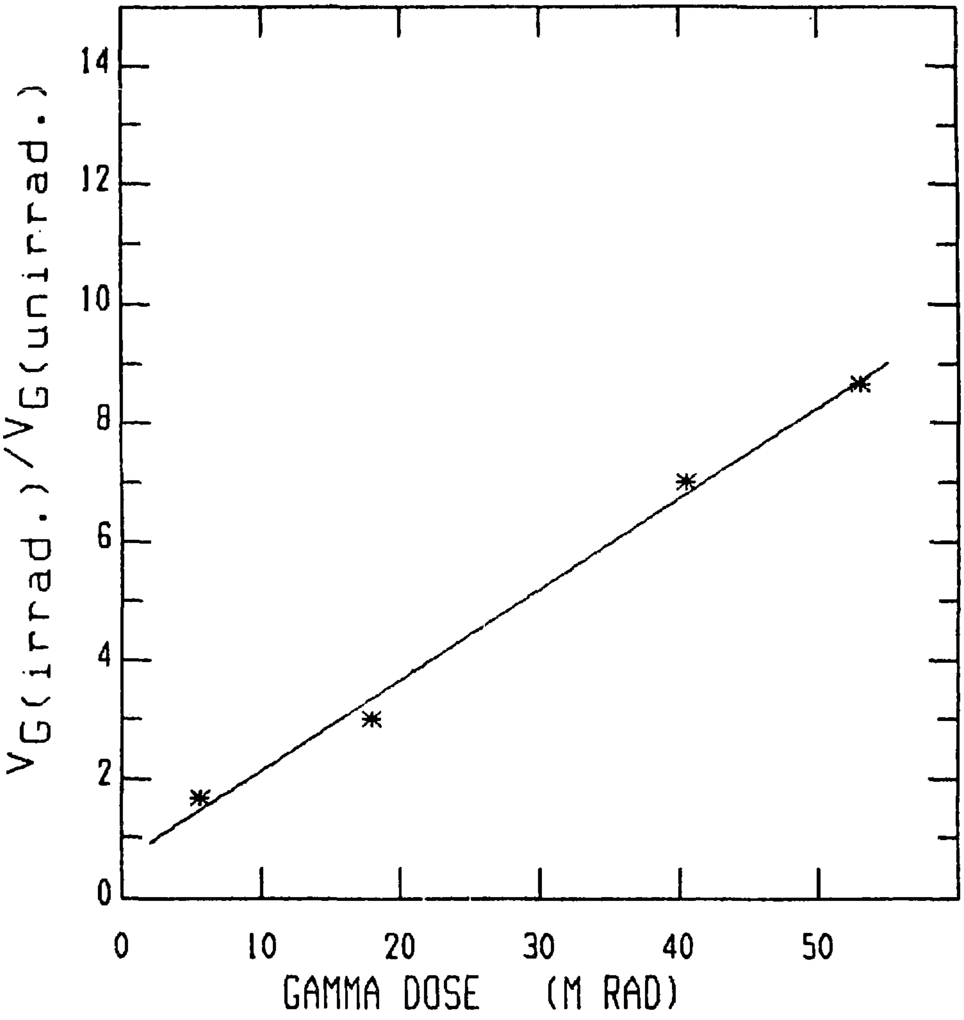

FIG.3b BULK ETCH RATE OF GAMMA IRRADIATED CR-39(DOP)

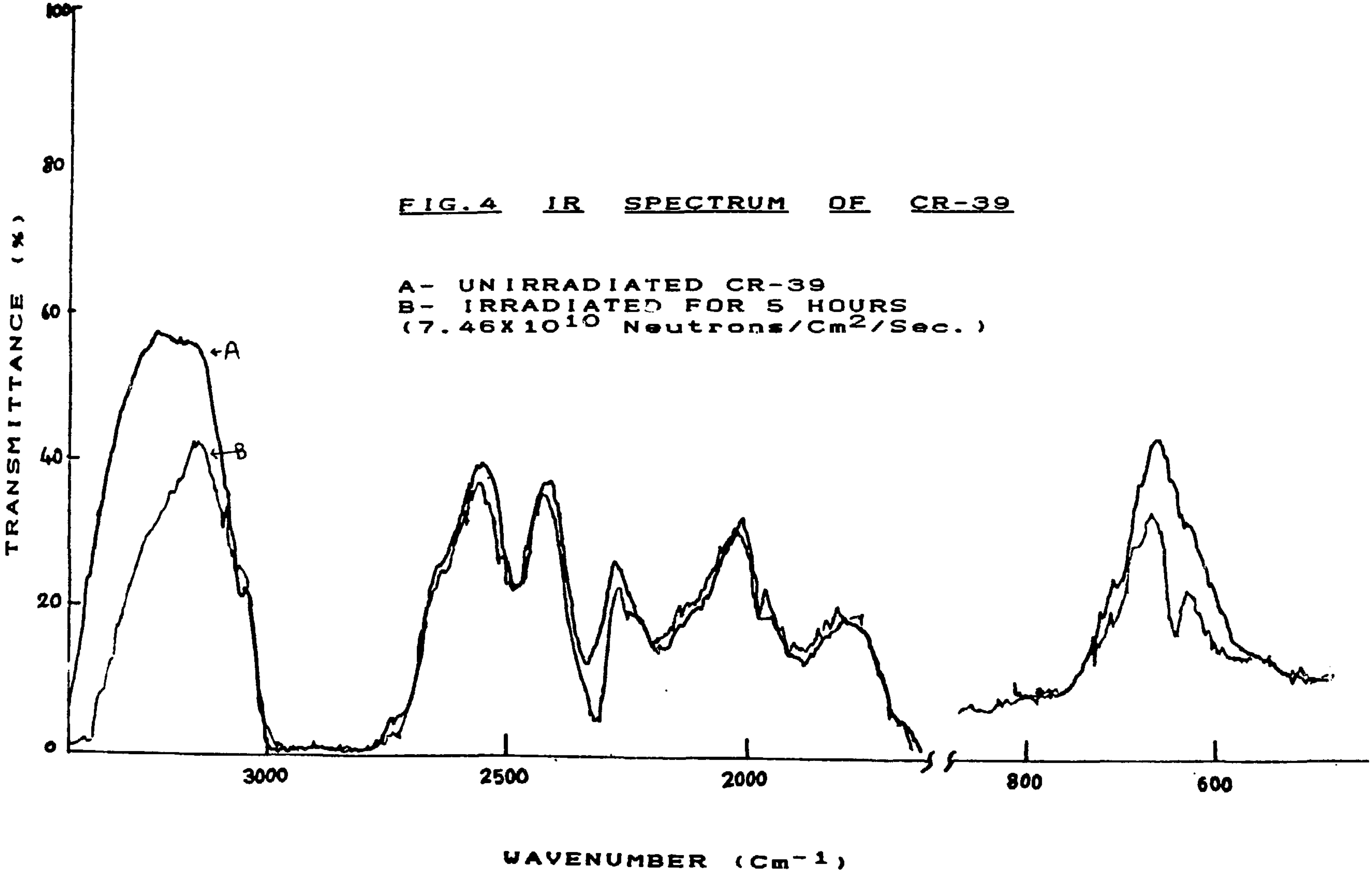

FIG.4 IR SPECTRUM OF CR-39

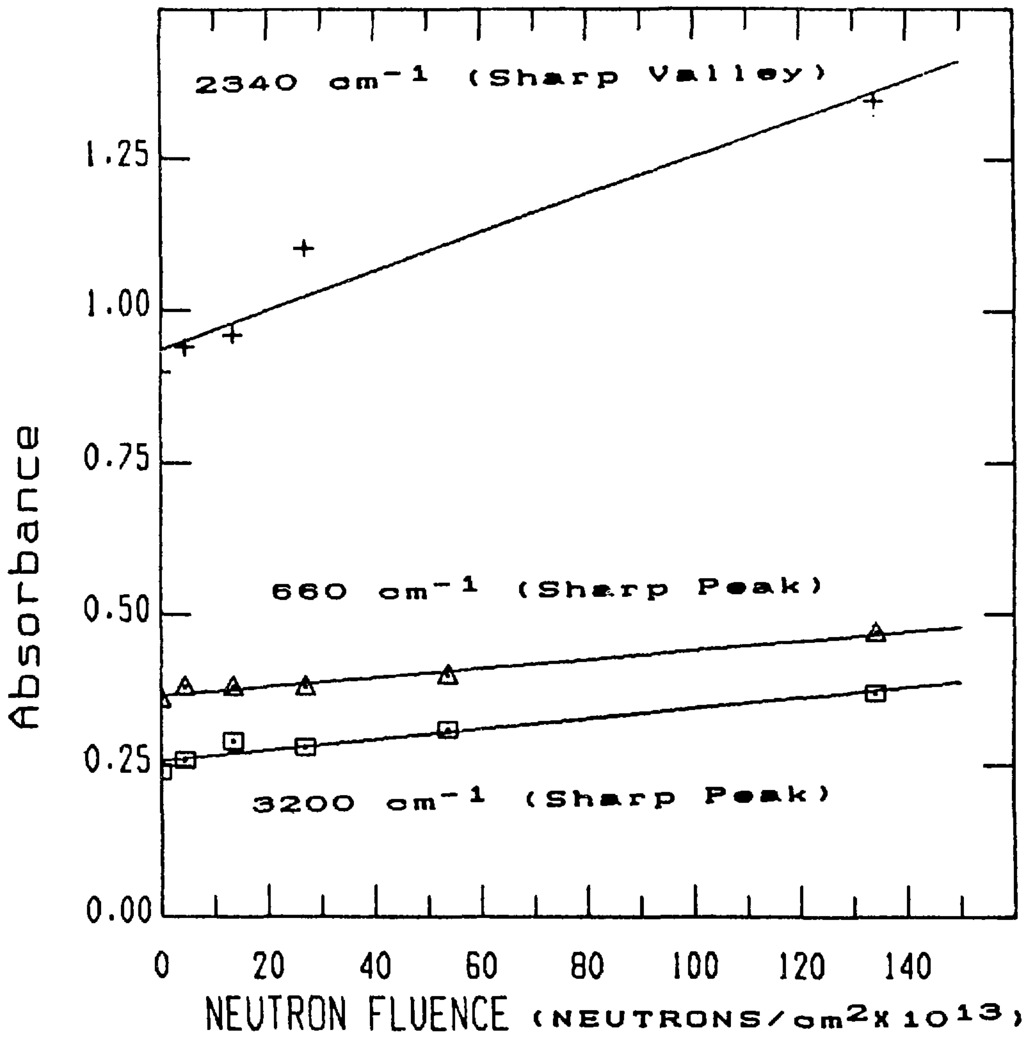

FIG.5 VARIATION OF ABSORBANCE WITH NEUTRON FLUENCE IN CR-39

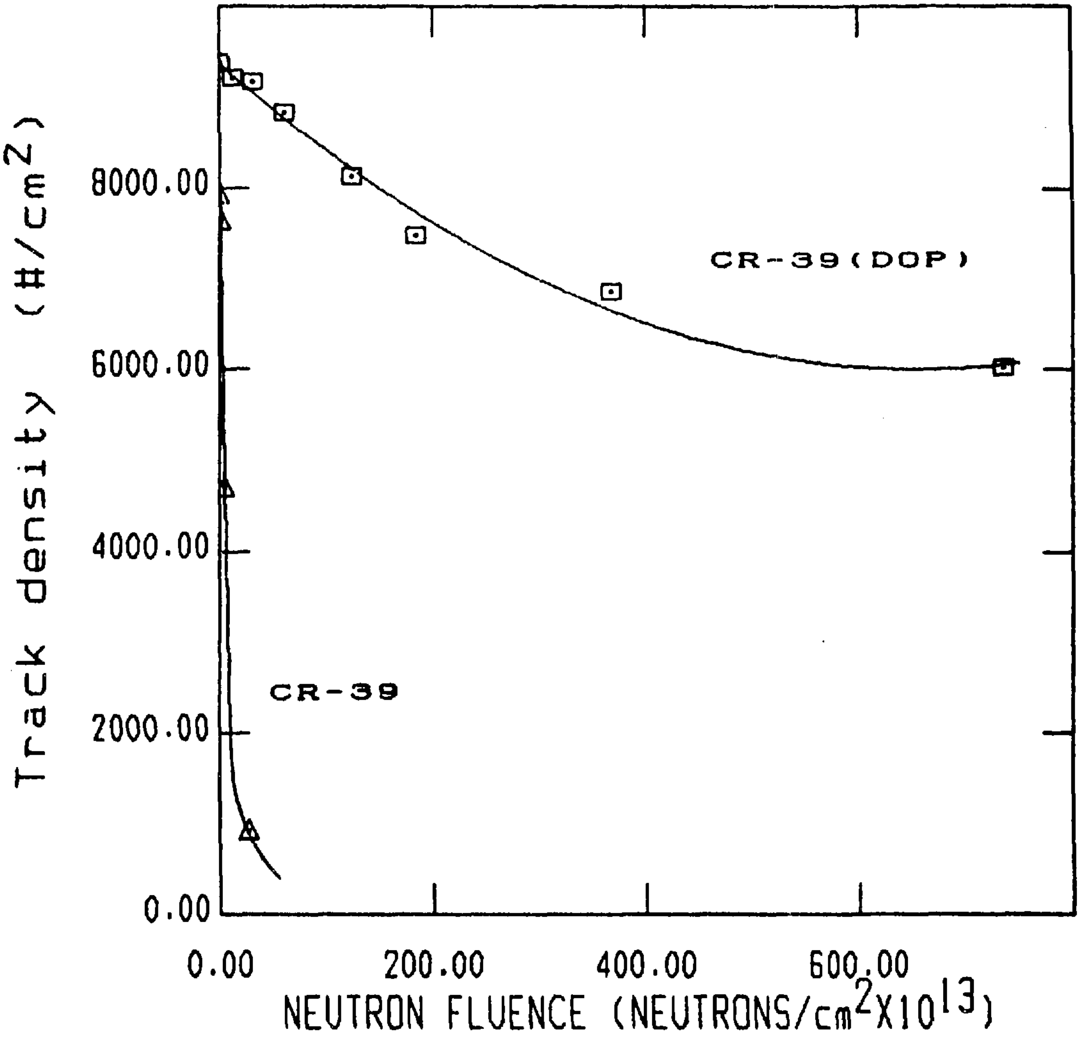

FIG.6 VARIATION OF FISSION TRACK DENSITIES WITH REACTOR NEUTRON FLUENCE IN CR-39 DETECTORS

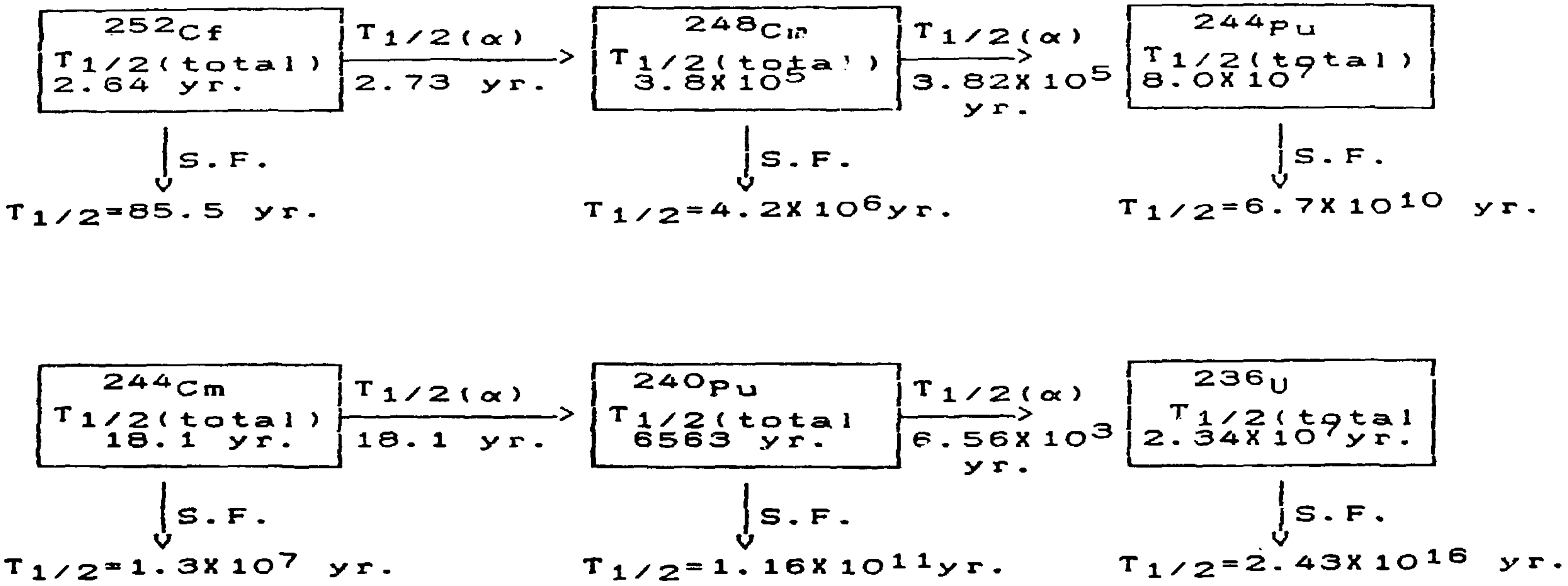

FIG.7 DECAY SCHEMES OF Cf^{252} & Cm^{244}

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