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Alpha Source Homogeneity Evaluation Using SSNTDs: A Comparative Study

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Abstract: Solid State Nuclear Track Detectors (SSNTDs) are known to reveal details about the impinging radiation such as energy, intensity etc. Analysis of irradiated SSNTDs has been attempted to study the effect of homogeneity of the alpha source used for irradiation. Comparative analyses of CN85 and CR39 detectors were carried out after irradiation with plutonium alphas. Image analysis and UV-Vis spectrophotometry were used for analysis.

Keywords: SSNTD, CN85, Alpha, Homogeneity

1. Introduction

Alpha detection using solid state nuclear track detectors (SSNTDs) is a very popular method for dosimetry, environmental surveys, geological studies etc [1-3]. Different types of polymer based SSNTDs such as cellulose nitrate, PADC, polycarbonate, polyethylene terphthalate are in use. These polymers are very sensitive to the energetic impinging of alpha particles. Impingement results in the intense damage trail and different properties of the alpha source can be obtained by analysing the etched tracks on the detector surface. Cellulose nitrate (CN) is a material used as SSNTD whose chemical formula is $C_6H_8O_9N_2$. CN85 is a commercially available Cellulose nitrate based SSNTD having density 1.33-1.6 gm/cm^3 and a foil thickness of 100-1000µm [4]. CR 39 is another commonly used SSNTD (Columbia Resin #39 - the 39th formula of a thermosetting plastic developed by the Columbia Resins project in 1940) which is a polyallyl diglycol carbonate with short polyallyl chains joined with links containing carbonates and diethylene glycol groups into a dense 3D network with an initiating monomer unit. It has a composition of $C_{12}H_{18}O_7$, density of 1.3 gm/cm^3 and a foil thickness of the order of 500 µm [5]. Interaction of radiations results in changes in the chemical structures and consequently in their physical and mechanical properties. It results in the formation of latent track which can be made visible by the process of chemical etching [6]. It is seen that the response of the SSNTDS are dependent on the etchant and the etching conditions used [7].

SSNTDs have been put into regular use in nuclear fuel fabrication facility for fuel characterization. Fuel characteristics such as composition and homogeneity can be quantitatively estimated using different analytical methodologies applied on the irradiated SSNTDs [8]. Plutonium is an alpha emitting nuclide which plays a crucial role in nuclear industry. The presence and distribution of Pu in mixed nuclear fuels can be monitored using alpha track analysis technique due to significantly high specific activity of plutonium. The process of imaging and analysis of the alpha emitting nuclide in a mixed nuclear fuel to estimate homogeneity is called alpha autoradiography. Conventionally the alpha images are manually evaluated with the help of optical microscope and image analytical methods applied for detailed analysis. UV-Vis spectroscopic studies revealed that changes in the spectral properties could be correlated to the alpha radiation intensity [9].

Homogeneity of the alpha source which is used for irradiation of the detectors was expected to reflect in the optical properties of the detectors. In this study, the nuclear

fuel samples chosen as alpha sources were prepared by different methods to ensure various levels of homogeneity and the changes of properties of irradiated SSNTDs viz CN-85 and CR-39 were evaluated and compared.

2. Materials and Methods

Plutonium is predominantly an alpha emitter with an average energy of 5.1 MeV. It has very high specific activity of 0.06Ci/g due to which it is viable to record its alpha image in presence of other alpha emitting nuclides of comparatively lower specific activity. Mixed oxide (MOX) nuclear fuels are synthesized to provide combination of a fissile material and fertile material in a fixed proportion as required for a specific reactor. Plutonium dioxide (PuO_2) is a fissile material (undergoes fission upon irradiation with neutrons of low energy) while thorium dioxide (ThO_2) is a fertile material (gets converted to a fissile material upon neutron irradiation). In this study, (Th, Pu)O_2 MOX fuel samples containing 1% PuO_2 has been used as alpha source for irradiating the SSNTDs.

Fuel samples have been synthesized by two different methods. The conventional powder metallurgy route (POP) involving mixing of oxide powders, milling, cold compaction and sintering was followed to synthesise pellets with homogeneous distribution of plutonium. Coated agglomerate pelletisation route (CAP) which involves extrusion and spherodization of the fertile material followed by coating with fissile material before cold compaction and sintering was customised to synthesise fuel samples having the same composition but low levels of homogeneity. The difference in homogeneity levels of the two samples were confirmed using XRD studies and then by alpha autoradiography using CR39 detector [8]. The two pellets were moulded together in Bakelite and sample surface preparation was carried out by grinding and polishing to ensure good contact with the detector surface.

Cellulose nitrate detector (CN-85 of thickness 100 µm, Kodak) was cut into 2×2 cm sized piece and arranged with aluminium mylar film of 11 µm thickness to minimise non perpendicular incidence of alphas. The detector was then irradiated with the fuel samples for 8 minutes inside an alpha tight glove box, decontaminated and subjected to chemical etching using 2.5 N NaOH at 60°C for 20 mins.

The CN85 film was examined through an optical microscope (LEICA-DM ILM) and the images grabbed using an attached digital camera for comparison. Selected frames from each image were analysed using an image analysis software (Metal power image analyser version 3.0.0.9 by Metal Power India (Pvt.) Ltd.). Area fraction of each frame covered with alpha tracks was estimated using the software. The spectrophotometric analysis of the detector was carried out using a high resolution UV–Vis JASCO V-650 double beam spectrophotometer supported by Spectra Manager TM II software. A schematic representation of the process of alpha imaging and analyses is shown in Figure 1.

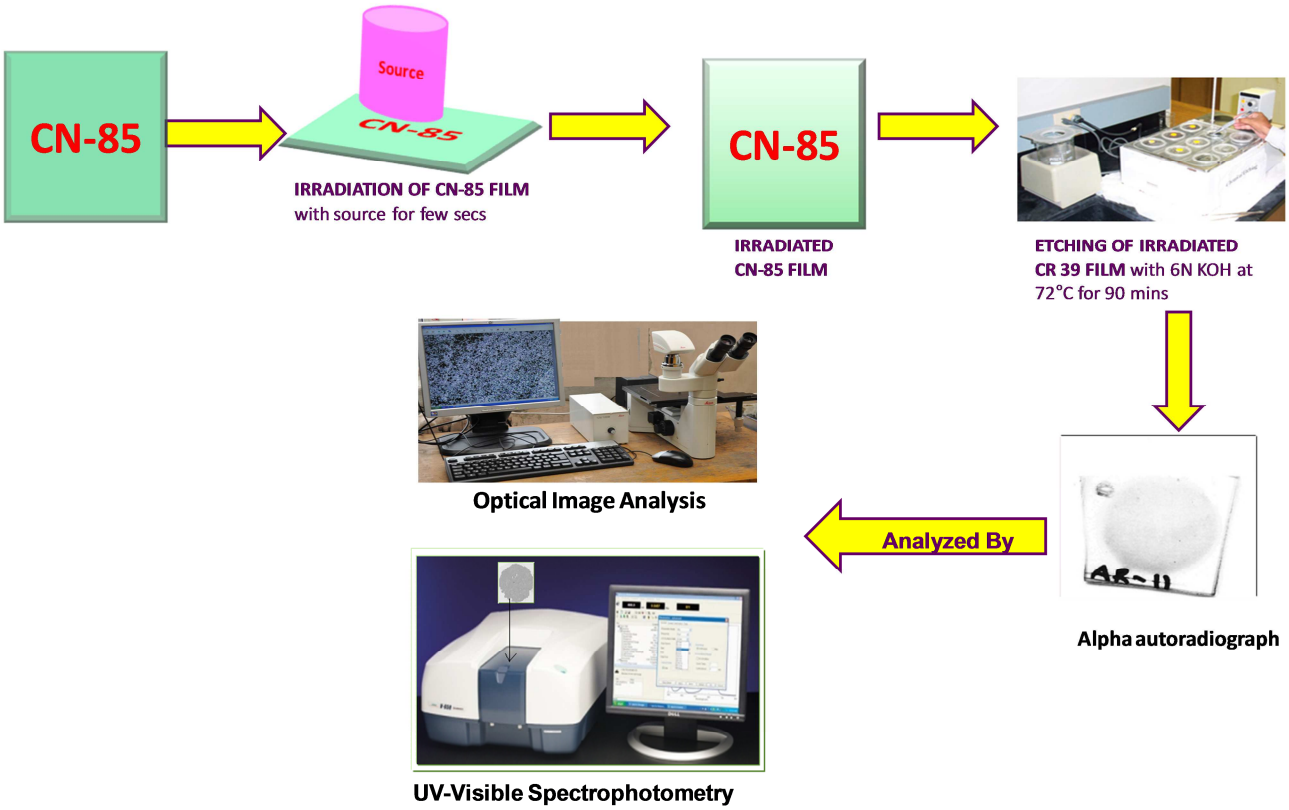

Figure 1. *Schematic of alpha imaging and analyses carried out in the study.*

3. Results and Discussion

The nuclear track formation is a complex phenomenon governed by different factors namely the nature and fluence of impinging radiation, detector material and etching parameters. This is reflected in the image characteristics of the tracks as well as the spectral features of the detectors [10].

3.1. Image Analysis

Selected frames of the microscopic alpha images of CAP and POP samples recorded on CN85 detector are shown in Figure 2 (a-d) and Figure 2 (e) respectively. Alpha images of the CAP sample revealed significant variation in track densities across the sample whereas the variation was only ±4.4% in case of the POP alpha image. Regions of very high as well as low track densities were observed in the CAP image as shown in Figure 2 (a). A number of frames were chosen manually and analysed so as to record the maximum and minimum track densities from each sample. Similarly, frames from the alpha autoradiograph corresponding to the maximum and minimum track densities of CAP sample and the image of POP sample both recorded on CR39 detector are shown in Figure 3 (a-b) and Figure 3 (c) respectively. The results of image analysis are given in Table 1 for comparison.

The variation of amount of PuO_2 (w%) obtained from alpha autoradiographic studies using CR39 was in the range of 0-3.9% in the CAP sample which was indicated by a variation of 1.4-25% in% irradiated area values obtained from image analysis. It is evident from Table 1 that CN 85 revealed a greater range of variation in% irradiated area values among the different frames of the alpha image corresponding to the heterogeneous source (CAP sample) as compared to CR 39.

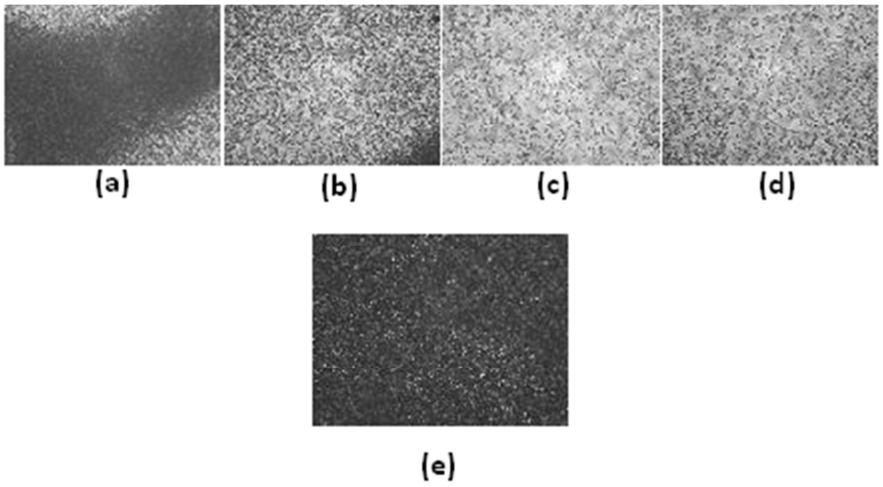

Figure 2. *(a-d) Alpha images of CN85 detectors irradiated with CAP and (e) POP samples.*

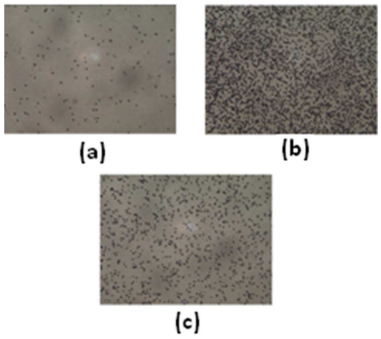

Figure 3. *(a-b) Alpha images of CR39 detectors irradiated with CAP and (c) POP samples.*

Table 1. *Results of Image Analysis of the Irradiated Detectors.*

Detector	% Irradiated Area using MOX fuel pellet as source	
	POP	CAP
CN-85	83.8	25.8
	91.4	14.0
	83.9	10.2
	85.6	12.9
	90.8	98.1
CR-39	10.9	1.42
	11.3	24.8
	10.6	22.9

3.2. UV Visible Spectrophotometry

The effect of alpha energy, fluence and etching period on the characteristic spectra of CR-39 detector has been illustrated earlier using standard alpha sources [9]. Alpha images of homogeneous (POP) and heterogeneous (CAP) sources were subjected to UV-Vis spectrophotometric analysis using a 10 mm aperture which was chosen so that the absorption of the full image portion on the detector could be recorded. The UV-Vis absorption spectra of CAP and POP samples recorded on CN85 detector are shown superimposed with blank portions of the detector in Figure 4 (a). A difference in characteristic absorption could be observed in the spectra even though both the samples were of same composition. This could be attributed to the difference in resultant Pu content in the contributing volume of the sample owing to heterogeneous distribution of the plutonium in the sample. Since plutonium alphas have a limited range of 18µm in the fuel matrix, the contributing volume will only have this thickness and hence the resultant Pu content of both the samples could be different. The effect of source homogeneity on spectral characteristics of CN-85 has been evaluated in the present study and compared with that of CR-39 [Figure 4 (b)].

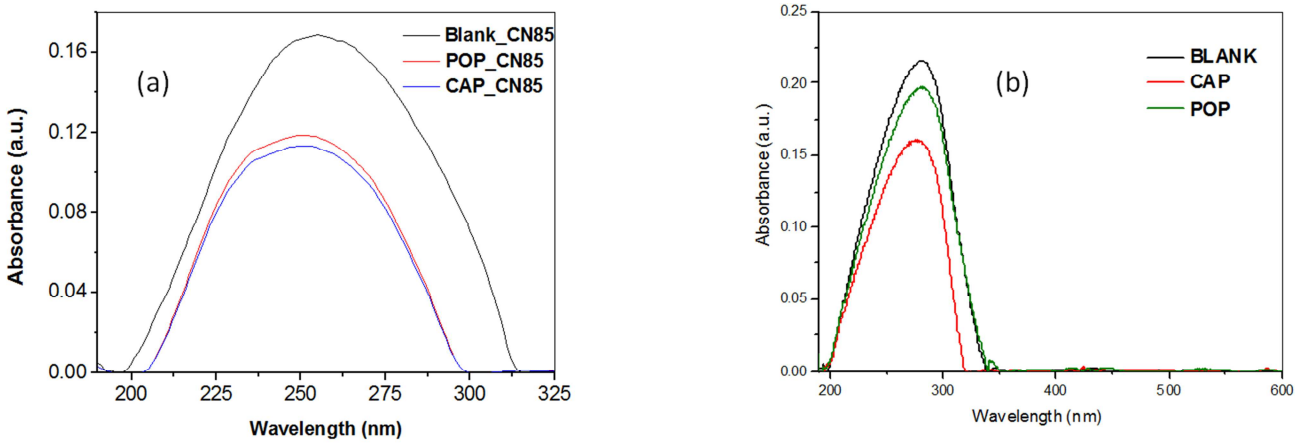

Figure 4. *UV-Visible absorption spectra of (a) CN85 and (b) CR39 irradiated with CAP and POP samples.*

Detailed investigation of source heterogeneity was carried out by recording the absorption spectra from smaller portions of each of the images with the help of a mini aperture of size 2mm. Each portion of the image corresponding to the CAP sample showed significantly different UV-Vis absorbance whereas that from POP image was insignificant. The spectra of CAP and POP recorded with mini aperture on CN85 are shown in Figure 5. The results of spectrophotometric analysis (UV-Vis) are given in Table 2.

From the studies, it is seen that the variation of UV-Vis absorbance values of different regions of CN-85 detector irradiated with CAP sample range from – 75% to +5% with respect to that of POP sample. The corresponding variation in our earlier studies using CR39 was – 23% to +10% [8].

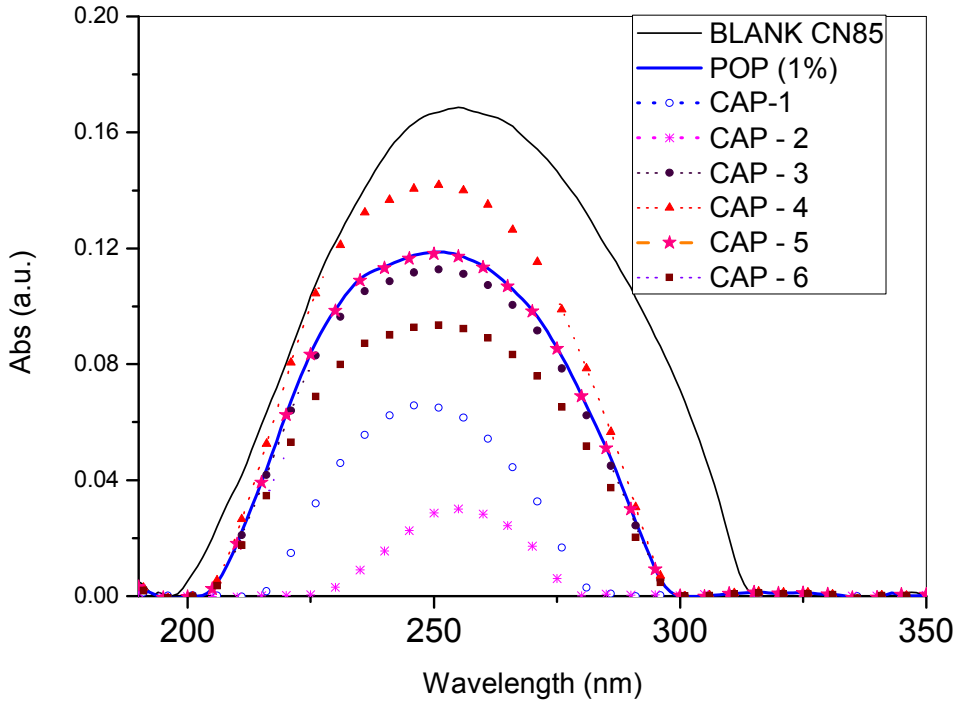

Figure 5. *Spectra of different portions of the a-autoradiograph.*

Table 2. *Results of spectroscopic analysis of the irradiated detector.*

Detector	Absorbance (a.u.)	
	POP	CAP
CN-85	0.118	0.139
		0.118
		0.112
		0.092
		0.065
		0.029

4. Conclusions

Response of CN85 detector revealed more details regarding homogeneity of plutonium bearing sources as compared to that by CR39 detector. The responses were identical in image analysis and UV-Vis spectrophotometric analysis of the detectors.

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