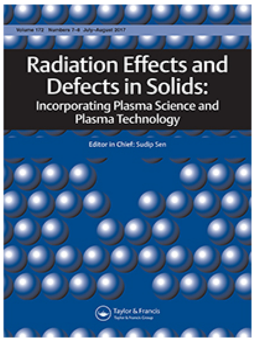

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# Track revelation and optical properties of pentaerythritol tetrakis (allyl carbonate) plastic for application as nuclear track detector: effects of gamma radiations

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**ABSTRACT**

The etching conditions of an indigenously prepared thin film of pentaerythritol tetrakis(allyl carbonate) (PETAC) were standardised for the use as a nuclear track detector. The optimum etching times in 6 N NaOH at 70°C for the appearance of fission and alpha tracks recorded in this detector from a $^{252}$Cf solid source were found to be 30 min and 1.50 h, respectively. The experimentally determined values for the bulk and track-etch rates for this detector in 6 N NaOH at 70°C were found to be $1.7 \pm 0.1$ and $88.4 \pm 10.7$ µm/h, respectively. From these results, the important track etching properties such as the critical angle of etching, the sensitivity and the fission track registration efficiency were calculated and compared with the commercially available detectors. The activation energy value for bulk etching calculated by applying Arrhenius equation to the bulk etch rates of the detector determined at different etching temperatures was found to be $0.86 \pm 0.02$ eV. This compares very well with the value of about 1.0 eV reported for most commonly used track detectors. The effects of gamma irradiation on this new detector in the dose range of 200–1000 kGy have also been studied using bulk etch rate technique. The activation energy values for bulk etching calculated from bulk etch rates measurements at different temperatures were found to decrease with the increase in gamma dose indicating scission of the detector due to gamma irradiation. The optical band gap of this detector was also determined using UV–visible spectrometry and the value was found to be $4.37 \pm 0.05$ eV.

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PETAC nuclear track detector; bulk- and track-etch rates; track registration efficiency; activation energy for bulk etching; UV–visible spectroscopy; optical band gap

## 1. Introduction

Solid-state nuclear track detectors (SSNTDs) are highly sensitive for recording rare radioactivity, energetic particle detections, neutron dosimetry, measurement of fission cross-sections in nanobarn range, etc. The particles tracks in SSNTD can also be developed as well defined through pores for the applications such as membranes and template for forming nanomaterials. In general, the plastics such as Lexan, Tuffak, CR-39, LR-115, Makrofol, Polyethylene Terephthalate, Cellulose Nitrate are used as SSNTD. Among these, the polyallyldiglycol carbonate (PADC) detectors (commercially available as CR-39 detectors) are

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extensively used as the SSNTD not only due to its high sensitivity towards energetic particles but also due to its optical properties after revelation of the particle tracks registered from harsh environment. In an effort to search a better polymer SSNTD, we have reported recently a synthetic protocol for preparing a thin film of pentaerythritol tetrakis (allyl carbonate) (PETAC) (*1*). The preliminary study has indicated that this material records fission as well as alpha tracks similar to CR-39. However, the parameters that control track revelation in SSNTDs such as the bulk etch ($V_b$) and track-etch ($V_t$) rates are important for the particle identification and quantification of the events recorded. The track-etch parameters are also important for track-shape-analysis-based automated slide scanner system (*2*). The chemistry-normality dependence of bulk etch rate in a CR-39 detector has been reported (*3*). The knowledge of $V_b$ for the nuclear track detectors at different temperatures is of considerable usefulness (*4*). It can also be used for the determination of activation energy for bulk etching and this is very often determined for nuclear track detectors (*4*–*7*). In the present work, the bulk etch rates of this newly developed material at different temperatures have been determined to understand its track revelation properties. The activation energy value for bulk etching was also determined by using Arrhenius equation and compared with the values given in the literature for the most commonly used nuclear track detectors. In addition to $V_b$, another important parameter called track-etch rate was also experimentally determined for this new polymer. From these results, the critical angle of etching, the sensitivity and the fission track registration efficiency were also calculated and compared with the commercially available detectors. The activation energy value for bulk etching and the optical band gap of this polymer were also determined and compared with the other detectors. As nuclear track detectors are also used in nuclear reactors for recording fission events, the gamma dose in the reactor environment is likely to change the properties of this detector as reported for the CR-39 and other polycarbonate detector (*6*, *8*, *9*). Therefore, it was thought interesting to study the gamma irradiation effects on this detector. The gamma irradiation effects in the present investigation were studied by using bulk etch rate method.

## 2. Experimental

Thin films of pentaerythritol tetrakis (allyl carbonate) (PETAC) were exposed to a planchetted $^{252}$Cf source in $2\pi$ geometry to register fission fragments as well as alpha particles. Etching of the exposed samples was carried out in different concentrations (5–7 M) of NaOH solution at different temperatures (60–80°C) and for different intervals of time (0.25–10 h). From the experiments, it was found that the optimum conditions for the etching of fission tracks in this polymer are 30 min etching time at 70°C in 6 M NaOH as the etchant. The images of fission tracks are displayed in Figure 1(a,b). In 15 min etching time, fission tracks start appearing and then they are fully developed in 30 min etching time. Under these conditions, alpha tracks are developed after 1 h 30 min of chemical etching. Although, the alpha tracks start appearing in 1 h 10 min but they are fully developed in 1 h 30 min. The images of alpha tracks are displayed in Figure 1(c,d). Experiments were also carried out to find out the bulk etch rate ($V_b$) and the track-etch rate ($V_t$) of this new polymer. The $V_b$ was determined by the gravimetric method (*10*). The polymer samples were etched in 6 M NaOH. The procedure for the gravimetric method involved immersion of a sample of the detector of known weight and thickness into the etchant solution, maintained at a fixed temperature

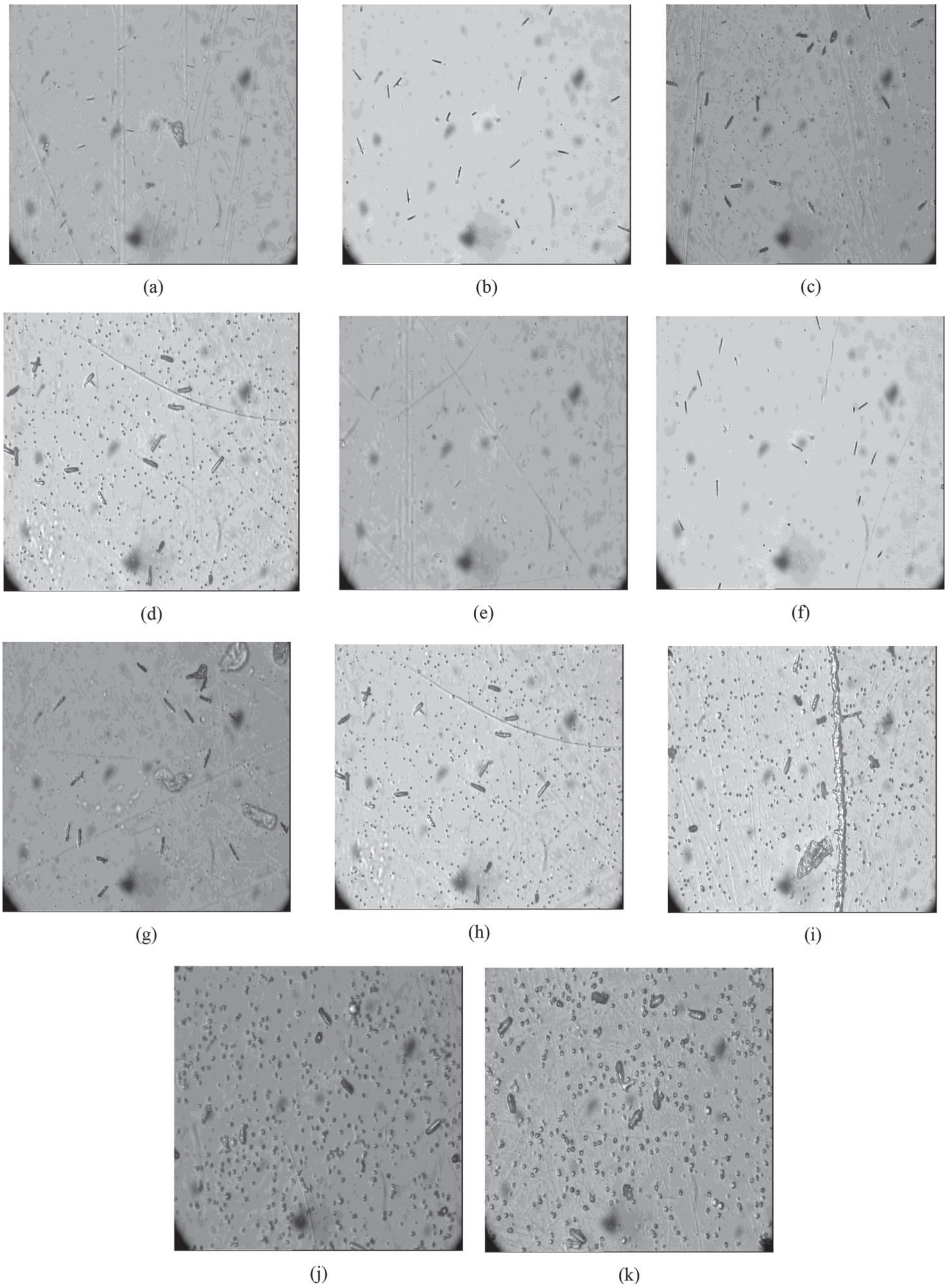
(a) (b) (c) (d) (e) (f) (g) (h) (i) (j) (k)

**Figure 1.** (a,b) Development of fission tracks in PETAC detector etched at 70°C for (a) 15- and (b) 30-min etching time in 6 N NaOH as the etchant. (c,d) Development of alpha tracks in PETAC detector etched at 70°C for (c) 1 h 10 min and (d) 1 h 30 min etching time in 6N NaOH as the etchant. (e–k) The image of fission tracks used to calculate $V_t$ at 70°C for (e) 15 min (f) 30 min (g) 70 min (h) 90 min (i) 120 min (j) 150 min (k) 180 min etching time in 6 N NaOH as the etchant.

in a thermostat, under conditions of stirring for an accurately noted period. It was taken out, thoroughly washed with demineralised water and was weighed again after air-drying at room temperature. $V_b$ was obtained in units of length per unit time as $V_b = mT_i/2\,\mathrm{M}t$, where $m$ is the weight (g) loss due to etching for a period, $t$ (h), $T_i$ is the initial thickness (μm) and $M$ is the initial weight of sample (g). The bulk etch rates were determined at different temperatures to deduce activation energy for bulk etching. For the track-etch rate measurements, the samples after exposing to a planchetted $^{252}$Cf source in $2\pi$ geometry were etched in 6 M NaOH at 70°C. The detectors were scanned for track length measurements after etching using a binocular microscope at a magnification of 40× and about 100 tracks were taken in each case. The average value of the track length corresponding to each etching interval was used for the track-etch rate ($V_t$) measurement (*11*). $V_t$ was calculated on the assumption that $V_t$ remains constant for very small etching times during which a small segment of the particle trajectory is etched out. The track-etch rate ($V_t$) was determined from the slope of the linear part of the curve between the average track lengths, $L$ and the etching time, $t$. The images of the fission tracks which has been used to calculate $V_t$ are displayed in Figure 1(e–k). From the bulk and track-etch rate measurements, the critical angle of etching, $\Theta_c$ ($\Theta_c = \sin^{-1} V_b/V_t$) and sensitivity, $S$ ($S = V_t/V_b$) were calculated. The fission track registration efficiency ($\eta$) was also calculated from $V_b$ and $V_t$ by using the following equation: etching efficiency, $\eta = 1 - \sin \Theta_c$ ; $\Theta_c = \sin^{-1} V_b/V_t$; therefore, $\eta = 1 - V_b/V_t$.

To find out the optical band gap of the new polymer, UV–visible spectra by a UV–VIS Spectrometer (Jasco, Model V-530) in the range of 200–800 nm of the samples were also recorded. To study the gamma irradiation effects on this new polymer, the polymer films were irradiated with $^{60}$Co gamma rays in air using the gamma irradiator of Radiochemistry Division, BARC (*6*). The samples were irradiated for various times and the total gamma doses obtained were 200.0, 400.0, and 1000.0 kGy.

## 3. Results and discussion

Table 1 gives the values of bulk etch rates determined at different temperatures for the unirradiated and the gamma-irradiated samples. The average values of $V_b$ and $V_t$ for this new polymer as determined by the above methods were found to be 1.7 ± 0.1 and 88.4 ± 10.7 μm/h, respectively, at 70°C in 6 M NaOH as the etchant. From these results, the critical angle of etching ($\theta_c = \sin^{-1} V_b/V_t$), sensitivity ($S = V_t/V_b$) and the fission track registration efficiency ($\eta = 1 - V_b/V_t$) were calculated and found to be 1.1°, 52 and 0.98, respectively. The fission track registration efficiency value of the present material compares well with the efficiency of commercial available materials such as Lexan, which is about 0.96 (*12*), Tuffak, which is about 0.97 (*13*), and CR-39, which is about 0.81 (*14*). For calculating the activation energy for bulk etching, the bulk etch rates were determined at different temperatures (Table 1). In Figure 2, we have plotted log $V_b$ against the reciprocals of the absolute temperatures for the unirradiated and gamma-irradiated polymer samples. These

**Table 1.** Gamma irradiation effects on activation energy for bulk etching of the new polymer [Pentaerythritol Tetrakis (Allyl carbonate) (PETAC) [Etching conditions: 6 N NaoH, 4 h]

| | $V_b$ (μm/h) at temperature | | | |
|---|---|---|---|---|
| Gamma dose (kGy) | 60°C | 70°C | 80°C | Activation energy for bulk etching, E (eV) |
| Unirradiated | 0.48 ± 0.04 | 1.70 ± 0.10 | 2.59 ± 0.13 | 0.86 ± 0.02 |
| 200.0 | 1.39 ± 0.11 | 1.45 ± 0.10 | 1.63 ± 0.10 | 0.40 ± 0.04 |
| 400.0 | 27.12 ± 1.36 | 31.66 ± 1.59 | 47.05 ± 1.88 | 0.30 ± 0.04 |
| 1000.0 | 29.25 ± 1.17 | 38.19 ± 1.92 | 53.91 ± 2.70 | 0.30 ± 0.03 |

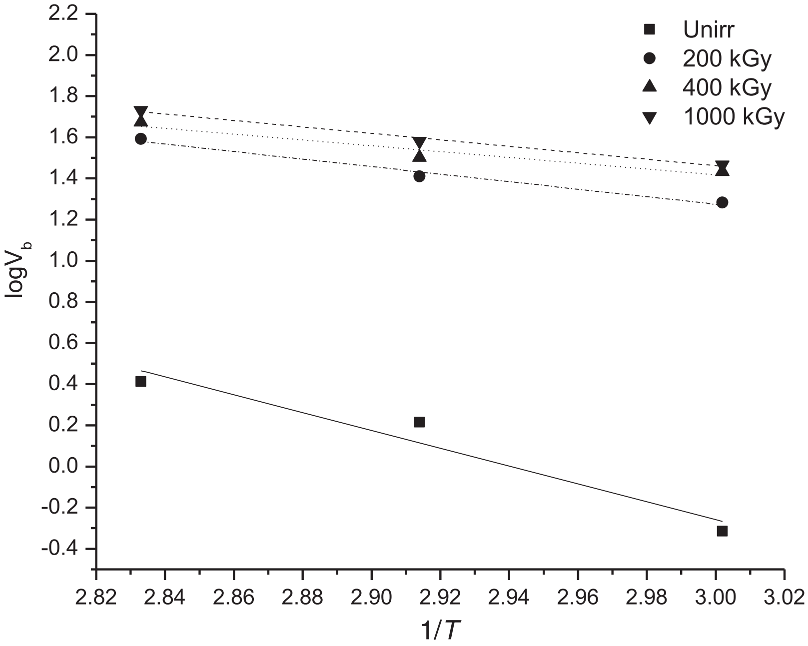

**Figure 2.** The plot of $\log_{10} V_b$ vs. $1/T$ for unirradiated and gamma-irradiated pentaerythritol tetrakis (allyl carbonate) (PETAC) homopolymer.

plots confirm to the expression $V_b = A \,.\, e^{-E/kT}$, where $E$ is the activation energy for bulk etching, $A$ is a constant for a given medium- etchant combination, $k$ is the Boltzmann constant and $T$ is the temperature in $K$. In Table 1, we have given the activation energy ($E$) values for the unirradiated and the gamma-irradiated samples. The $E$ value for the unirradiated sample is found to be 0.86 ± 0.02 eV and it is given in Table 2 along with the $E$ values reported in the literature for some most commonly used nuclear track detectors (*4*, *6*, *7*, *10*, *15–17*). There seems to be good agreement between the value obtained for this new polymer and the values reported for the most commonly used track detectors. Recently, the activation energy values for the bulk etching for different SSNTDs have also been reviewed by Hermsdorf (*18*). The majority of the experimentally determined values are found to be in the order of 0.8 ± 0.2 eV. Such large differences in the values indicate that the activation energies depend on the etchant concentration too. The activation energy values for the gamma-irradiated samples have been found to decrease with the increase in gamma dose. This decrease in activation energy values indicates the chain scission of the polymeric molecules by gamma rays (*6*, *15*, *16*, *19*). The effects of gamma rays on the activation energies of the polymers have also been discussed by Hermsdorf in his latest review (*18*). The materials modifications due to gamma rays result here in reduced activation energies for bulk etching. This indicates faster etching processes. However, drastic changes in the track reveling property of gamma exposed polymer material have been observed for gamma ray doses above $10^5$ Gy only. In the present investigation, we have also observed the major reduction in activation energy value for gamma-irradiated PETAC samples in the dose range of 200–1000 kGy. Such studies are very important because SSNTDs will be favourably applied in mixed radiation fields appearing in fission and fusion reactor facilities

**Table 2.** Value of the activation energy for bulk etching of the new polymer [Pentaerythritol Tetrakis (Allyl carbonate) (PETAC) along with the values reported in the literature for the commonly used nuclear track detectors.

| Nuclear track detector | Activation energy for bulk etching, *E* (eV) |
|---|---|
| PETAC | 0.86 ± 0.02 |
| ABNEC:ADC (1:9) | 0.98 ± 0.07 |
| Lexan | 0.75 ± 0. 04 |
| Cellulose acetate | 0.95 ± 0. 01 |
| Lexan | 0.74 ± not quoted |
| Polyester | 0.94 ± not quoted |
| Makrofol KG | 0.99 ± 0.04 |
| Makrofol KL | 1.18 ± 0.08 |
| Makrofol N | 0.89 ± 0.04 |
| CR-39 (Pitsburgh) | 0.85 ± 0.05 |
| CR-39 (Type unknown) | 0.92 ± not quoted |
| CR-39 (Homalite) | 0.89 ± 0.04 |
| CR-39 (Homalite) | 0.83 ± 0.04 |
| CR-39 (Pershore) | 0.78 ± 0.03 |
| CR-39 (Pershore) | 0.79 ± 0.02 |
| CR-39 (Pershore) | 0.77 ± 0.02 |
| CR-39 (Pershore) | 0.79 ± 0.02 |
| NADAC:ADC | 0.93 ± 0.07 |
| CR-39 (Tastrak) | 0.57 ± 0.02 |
| CR-39 (Italian) | 0.75 ± not quoted |
| CR-39 (Hungarian) | 0.72 ± not quoted |

or cosmic and space research (*18*). The UV–visible absorption spectra of the unirradiated material in the wavelength range 260–500 nm is presented in Figure 3. The optical band gap of this new polymer was calculated from the optical wavelength data and is found to be 4.37 ± 0.05 eV. This value is reported in Table 3 along with the values reported for some other track detectors in the literature. The optical band gaps for most of the commonly used track detectors are found to be in the range of 4.16–4.44 eV (*15*, *16*, *19*–*22*).

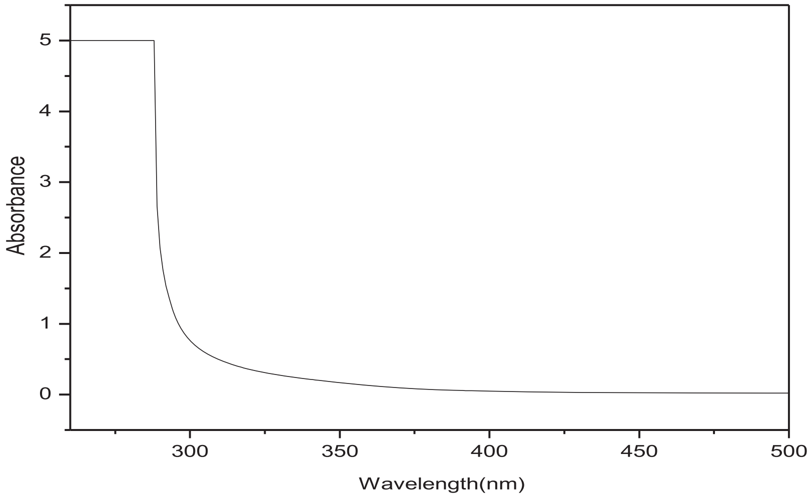

**Figure 3.** UV–vis spectra of unirradiated PETAC homopolymer.

**Table 3.** Value of the optical band gap ($E_g$) of the new polymer along with the values reported in the literature for the most commonly commercially available polymers.

| Polymer | Optical band gap $E_g$ (eV) |
|---|---|
| PETAC | 4.37 (Present work) |
| CR-39 [Pershore] | 4.40 (16) |
| Tuffak Polycarbonate | 4.40 (19) |
| NADAC – ADC Copolymer | 4.23 (15) |
| Lexan Polycarbonate | 4.44 (20) |
| Makrofol KG | 4.44 (21) |
| Makrofol N | 4.44 (21) |
| CR-39 [Tastrak] | 4.16 (22) |

## 4. Conclusions

The indigenously prepared polymer pentaerythritol tetrakis (allyl carbonate) (PETAC) is found to be a good substitute as a solid-state nuclear track detector to the imported lexan, Tuffak, CR-39, etc. for recording tracks. The fission track registration efficiency, the activation energy for bulk etching and the optical band gap of this material compares very well with some of the most commonly used imported track detectors such as Lexan polycarbonates and CR-39., thus making it an ideal import substitute particularly for recording fission tracks. The gamma irradiation effects studied on this new film in the dose range 200.0–1000.0 kGy by the bulk etch method indicate scission of the film due to gamma irradiation.

## Disclosure statement

No potential conflict of interest was reported by the authors.

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## References

(1) Mandrekar, V.K.; Chourasiya, G.; Kalsi, P.C.; Tilve, S.G.; Nadkarni, V.S. *Nucl. Instrum. Methods B* **2010,** *268,* 537–542.
(2) Bátor, G.; Csordás, A.; Horváth, D.; Somlai, J.; Kovács, T.A. *J. Radioanal. Nucl. Chem.* **2015,** *306,* 333–339.
(3) Gupta, S.K.; Singh, P.; Kumar, R.; Kumar, S. *Adv. Polym. Technol.* **2015,** *34,* 21510 (6).
(4) Dwivedi, K.K.; Mukherji, S.M. *Nucl. Instrum. Methods* **1979,** *159,* 433–438.
(5) Mhatre, A.; Chavan, V.; Kalsi, P.C.; Nadkarni, V.S. *J. Radioanal. Nucl. Chem.* **2010,** *286,* 99–101.
(6) Agarwal, C.; Kalsi, P.C.; Ramaswami, A. *Radiat. Eff. Def. Solids* **2006,** *161,* 131–133.
(7) Nikezic, D.; Yu, K.N. *Mater. Sci. Eng. R* **2004,** *46,* 51–123.
(8) Sharma, T.; Aggarwal, S.; Sharma, A.; Kumar, S.; Mittal, V.K.; Kalsi, P.C.; Manchanda, V.K. *Radiat. Eff. Def. Solids* **2008,** *163,* 173–179.
(9) Al-Jubbori, M.A. *Appl. Radiat. Isot.* **2016,** *118,* 228–231.
(10) Kalsi, P.C.; Nadkarni, V.S.; Manchanda, V.K. *Radiat. Phys. Chem.* **2008,** *77,* 1002–1004.
(11) Singh, S.; Prasher, S. *Nucl. Instrum. Methods B* **2004,** *222,* 518–524.
(12) Iyer, R.H.; Chaudhuri, N.K. *Radiat. Meas.* **1997,** *27,* 529–548.
(13) Kalsi, P.C. *J. Radioanal. Nucl. Chem.* **1999,** *242,* 169–172.

(14) Bhanu, A.U.; Iye, R.H. *Nuclear Tracks: Application to Earth Science and Nuclear Physics*, Sharma, K.K., Ed.; Organizing committee of the fourth National Seminar-cum-Workshop on SSNTD: Dehradun, 1986; 181.

(15) Chavan, V.; Kalsi, P.C.; Mhatre, A. *J. Radioanal. Nucl. Chem.* **2011,** *287,* 273–276.

(16) Mhatre, A., Kalsi, P.C.; Choudhary, O.S.; Sachdev, G.P. *J. Radioanal. Nucl. Chem.* **2011,** *288,* 795–798.

(17) Hermsdorf, D.; Hunger, M.; Starke, S.; Wieckert, F. **2007,** *42,* 1–7.

(18) Hermsdorf, D. *Radiat. Meas.* **2012,** *47,* 518–529.

(19) Sapkal, J.A.; Kalsi, P.C.; Agarwal, C.; Thanamani, M.; Murali, S. *Radiat. Phys. Chem.* **2009,** *78,* 81–84.

(20) Singh, S.; Prasher, S. *Radiat. Meas.* **2005,** *40,* 50–54.

(21) Singh, S.; Prasher, S. *Radiat. Eff. Def. Solids* **2004,** *159,* 359–367.

(22) Sharma, T.; Aggarwal, S.; Kumar, S.; Mittal, V.K.; Kalsi, P.C.; Manchanda, V.K. *J. Mater. Sci.* **2007,** *42,* 1127–1130.