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Research Article

Effect of Nuclear Radiation Generated Thermal Spike on Degradation Reaction Kinetics of a Polymer SSNTD and Its Impact on Track Diameter

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Abstract. It has been extensively studied that high polymer form an important group of materials used for solid state nuclear track detectors (SSNTD). The radiation induced damage not only leave tracks for microscopic examination but also change other physical properties of the material. One of them being the average molecular mass of the polymer. The change in this parameter which is induced by the radiation exposure can be studied and can give an estimate of the number of polymer chain scission, which has a direct influence on the track diameter and can give an estimate of the energy of radiation deposited inside the sample. From this the energy of the incoming particle flux can be calculated. In this paper a relationship between polymer degradation as given by the extent of the depolymerization reaction, the track diameter and energy of the incoming particle is sought.

Keywords. SSNTD, Thermal Spike model, Polymers, Solid State reaction kinetics

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1. Introduction

The thermal spike model of Seitz and Koehler [8] has been used as a mechanism by which considerable damage to the target material by the nuclear radiation is produced. In this model the passage of energetic particle produces intense heating along the particle track which leads to cleavage of intermolecular bonds in the target material. This idea has been extended by Bonfiglioli *et al*. [1] and by Chadderton and co-workers [2–4]. Recently, Szenes (*et al*.) [9–11] has extensively studied this model in context of track formation by swift heavy ions in insulators and semiconductors. The thermal spike model is derived from considerations of classical law of heat conduction and is described in the subsequent section. When a high intensity nuclear radiation is impinged on a polymer SSNTD, the track formation results in the breakage of polymer chain at random locations. This reduces the molecular weight of the polymer. The polymer degradation can be modeled by a basic kinetic equation which expresses the reaction rate as a function of temperature (see for example Vyazovkin and Wigth [12]). Thus there is a direct connection between the sudden temperature enhancement due to a thermal spike and the extent of degradation in the detector material and will be explored in this paper.

2. The Thermal Spike Model

Energetic heavy ions interact very strongly with the electrons of the material in which they penetrate. If a particle deposits an energy Q per unit length at time $t = 0$, then the considerations of the classical law of heat conduction shows that the temperature T as a function of time t and the radial distance from the axis of the ion path is given by (Durrani and Bull [6]):

$$T(r,t) = T_0 + \frac{Q}{4\pi cd}\frac{1}{Dt}e^{-r^2/4Dt}, \tag{2.1}$$

where T_0 is the initial temperature, c is the heat capacity of the medium, d is the density and $D = \frac{\sigma}{cd}$, where σ is the thermal conductivity of the medium. A thermal spike will result in raising the temperature of the narrow cylindrical region of the medium by several thousand of K for short duration of time. When this energy flows through the material via thermal conduction, the temperature in the bulk material is raised for a longer duration. This results in considerable degradation of the polymer material over the entire available volume within the track.

3. Kinetics of Thermal Degradation of Polymers

Thermal degradation of polymers has been extensively studied in other contexts. The kinetics of polymer degradation are usually described by the basic kinetic equation, e.g., equation given by Flynn [7]:

$$\frac{d\alpha}{dt} = k(T)f(\alpha), \tag{3.1}$$

where α is the extent of the reaction and its value ranges from 0 to 1, t is the time, $k(T)$ is the rate constant and $f(\alpha)$ is the reaction model, which describes the dependence of reaction rate on the extent of reaction. In most cases the temperature dependence of the rate constant can be accurately described by the Arrhenius equation:

$$k(T) = A\exp\left(\frac{-E}{k_B T}\right), \tag{3.2}$$

where A is the preexponential factor and E is the activation energy.

4. The Proposed Model

We assume that an energetic particle impinges on the surface of a target material having cylindrical geometry. The radius of the target material is R_0 and the length is l_0. We assume that the energetic particle produces a track along the axis of the cylinder. We pick a cylindrical shell of radius R, where R can be anywhere between 0 and R_0. The thickness of the cylindrical shell is dR. We study the changes occurring in the material along this shell. The temperature in this shell will be uniform at all points on this shell as is ascertained by the radial symmetry of eq. (2.1). Substituting eq. (2.1) in eq. (3.1) we get:

$$\frac{d\alpha}{dt} = A\exp\left(-\frac{E}{k_B\left(T_0 + \frac{Q}{4\pi cd}\frac{1}{Dt}e^{\frac{-R^2}{4Dt}}\right)}\right)f(\alpha) \tag{4.1}$$

which leads to an integral equation:

$$\int_0^{\alpha}\frac{d\alpha}{f(\alpha)} = \int_0^{t} A\exp\left(-\frac{E}{k_B\left(T_0 + \frac{Q}{4\pi cd}\frac{1}{Dt}e^{\frac{-R^2}{4Dt}}\right)}\right)dt. \tag{4.2}$$

The limits are chosen such that the reaction proceeds within the corresponding time range in which the decomposition of the material is allowed. As a first guess one can take the time range of the order of the cooling time constant $\tau \sim mc/\sigma A$, where m is the mass of the reactive region inside the target, c is the heat capacity, σ is the thermal conductivity and A is the surface area of the cylindrical shell.

Taking the first derivative of eq. (2.1) with respect to t and equating it to 0 gives us the value of the time at which the maximum temperature is attained. The value of the maximum temperature is also obtained by this method. Simple calculation shows that the time at which maximum temperature is attained is given by:

$$t_m = \frac{R^2}{4D} \tag{4.3}$$

and the corresponding maximum temperature is:

$$T_m = T_0 + \frac{4Q}{4\pi cdR^2 e} \tag{4.4}$$

Taking the standard values of thermal conductivity and heat capacity for Lexan polymer which is a polycarbonate, one calculates the value of T_m for $Q = (dE/dx) * d$, where dE/dx is equal to 20000 MeV.cm^{2}g^{-1} for Ar-40 ion.

The value of T_m that we obtain is of the order of 10000 K for radial distance of the order of 2.80 nm. The time taken to reach this temperature is of the order of 1.5×10^{-11}s. On the other hand, at $R = 4.77$ nm, the maximum temperature reached is of the order of 3500 K. The time taken to reach this temperature is 4.37×10^{-11}s.

5. Results and Discussion

We plot the reaction profile of the polymer degradation caused by nuclear radiation as a function of radial distance from the track axis. We used eq. (4.2) to calculate the extent of the reaction as the function of the radial distance by integrating the equation using Simpson 1/3 rule. We assume a Gaussian spike peaked at temperature T_m and a width of the order ofτ where τ is the cooling time constant. The quadrature is calculated by using a 3 point Simpson's rules, the three points being $t = 0$, τ and 2τ. The values of A and E are taken from [5]. The extent of the reaction is calculated as a function of radial distance R and the amount of energy deposited per unit length Q. The results are plotted in Figure 1 for two values of Q. Only a slight difference by a factor of 1.2 in the Q value results in a measurable difference in the R values.

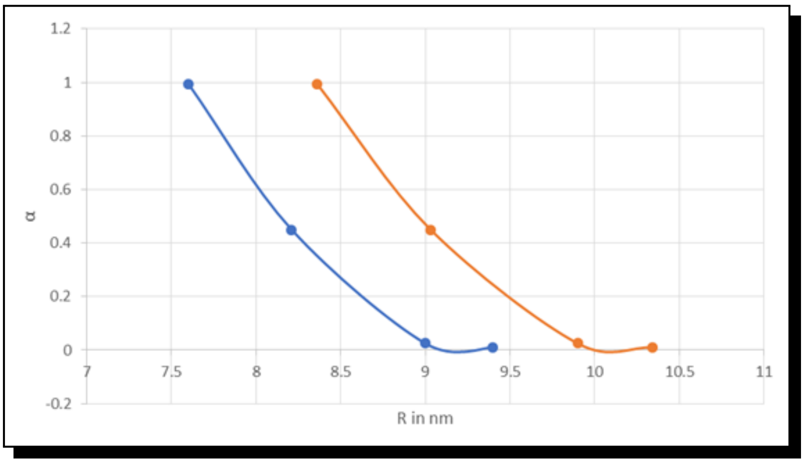

Figure 1. The extent of depolymerization reaction as a function of radial distance from the track axis. First order reaction mechanism is considered

For comparison the extent of depolymerization vs radial distance for second order and third order kinetic model are shown in Figure 2 and 3, respectively. The values of kinetic parameters are taken from [5]. The Q value is taken to be identical to the value taken in Figure 1.

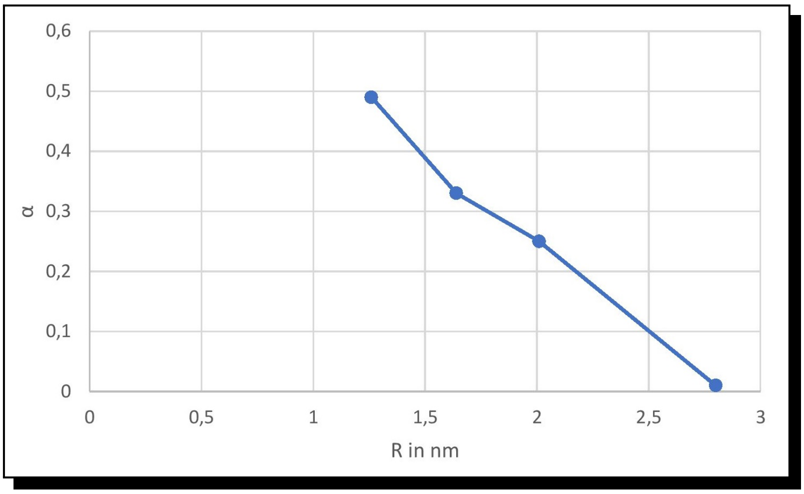

Figure 2. Extent of reaction vs radial distance for second order kinetic model of polymer degradation

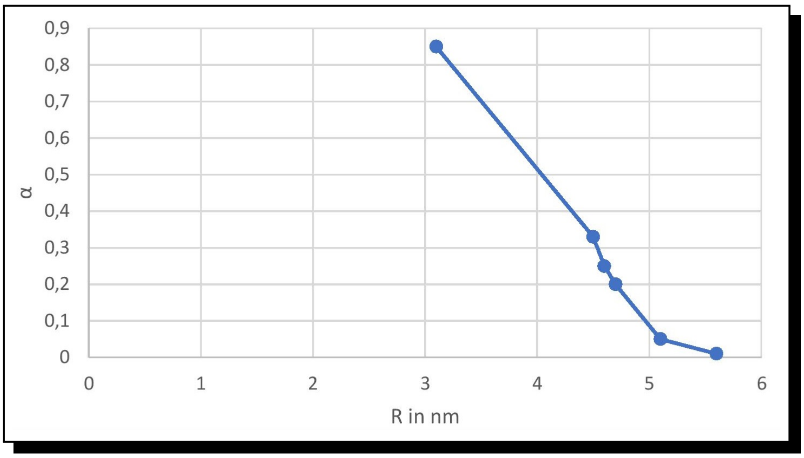

Figure 3. Extent of reaction vs radial distance for third order kinetic model of polymer degradation

As can be seen from the three above shown plots, the track diameter is significantly dependent on the reaction kinetics model being followed by the polymer during decomposition. The track diameter is maximum if a first order kinetics is followed. It is slightly less for third order kinetics and minimum for the second order kinetics. This dependence on reaction model is a result of the interplay of various parameters like the preexponential factor, the activation energy along with the power law obeyed by different reaction models. Thus, the experimental observation on track diameter can also give important information regarding the reaction mechanism involved in polymer degradation due to incident radiation, since the value of the track diameter is dependent on the order of the reaction being followed.

As can be seen from Figure 1, there is a sharp fall in the extent of the decomposition reaction within a range of 7.5 nm to 9.0 nm. For the values of R less than 7.5 nm the reaction essentially completes and the material evaporates completely. On the other hand, for values of R greater than 9.0 nm there is no change in the material chemistry and the material remains intact. From the electron microscopic examination of track diameter one can therefore get a knowledge of the value of Q as it depends significantly on this diameter. As can be seen from Figure 1, the shape of the graph for different values of Q remains almost the same but the track radial dimension shifts drastically with the change in the energy of the incoming radiation.

6. Conclusion

The Q value deduced from observations on track diameters can indirectly give information on the stopping power of the medium for a particular radiation. Using the value of the stopping power, one can apply Bethe-Bloch formula to infer the energy of the incoming particle if the ionic species be known. The reaction equations and hence the model are applicable to any solid state material being used as nuclear track detectors. Thus, the procedure described in this paper can lead to polymer SSTND becoming an important analytical tool in the field of nuclear radiation spectrum studies.

Competing Interests

The authors declare that they have no competing interests.

Authors' Contributions

All the authors contributed significantly in writing this article. The authors read and approved the final manuscript.

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