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Technical notes

Thermodynamics of nuclear track chemical etching

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A B S T R A C T

This is a brief paper with new and useful scientific information on nuclear track chemical etching. Nuclear track etching is described here by using basic concepts of thermodynamics. Enthalpy, entropy and free energy parameters are considered for the nuclear track etching. The free energy of etching is determined using etching experiments of fission fragment tracks in CR-39. Relationship between the free energy and the etching temperature is explored and is found to be approximately linear. The above relationship is discussed. A simple enthalpy–entropy model of chemical etching is presented. Experimental and computational results presented here are of fundamental interest in nuclear track detection methodology.

1. Introduction

The damage trails created by a charged particle penetration into a solid are preserved in nuclear track detectors and show preferential chemical etching [1–7]. Nuclear track etching is an essential step in nuclear track detection technique (NTDT). Understanding characteristics of nuclear track etching will enhance applications of the NTDT in various fields of nuclear science and technology. Thermodynamics can be used to understand and predict behavior of materials under processing. This short paper presents thermodynamics of nuclear track etching in CR-39 nuclear track detector using basic thermodynamic concepts and their calculations from experimental observations.

2. Experiments

CR-39 detector sheets of 1000 μm thickness (from Pershore Mouldings Ltd., UK) were used to prepare detector pieces, each of 2.5×2.5 cm^2 area. The prepared detectors were examined through the optical microscope to select detectors with clean and defect free surface. Selected detectors were exposed with fission fragments of ^{252}Cf. The irradiation experiments were performed in 2π-geometry in which detectors were directly placed on the fission fragment source. Used ^{252}Cf radiation source was of known strength and exposure time was adjusted to avoid the overlap of tracks after etching (keeping the track density up to 10^4 tracks/cm^2). CR-39 detectors, exposed to fission fragments, were chemically etched in the NaOH aqueous solution. The concentration or normality of the etching solution varied from 3N to 7N while temperature varied from 60 to 80 °C. Diameters and lengths of fission fragments tracks in etched CR-39 detectors were measured after each etching step in NaOH solution using Leitz DAILUX 22EB and ZEISS optical microscopes with the magnification of 400–1000. The observed track depths in CR-39 were multiplied with the refractive index of CR-39, which is 1.5. Fig. 1 is a schematic showing important aspects of nuclear track chemical etching experiments with essential information in the caption.

3. Results and discussion

Fig. 2 shows the mean fission fragment track lengths observed under different etching conditions. Experimental details are given in previous studies [8–10]. Fig. 3(a) shows the bulk etch rates of CR-39 under different etching conditions. Fig. 3(b) illustrates the track etch rates of fission fragments in CR-39 measured under the same etching conditions. Error analysis includes both systematic and statistical errors. Sources of errors include uncertainties in the set etching temperature and concentration of the etching solution. Microscopic measurement of tracks also introduces error in the observed parameters due to the instrumental limitations including the least count. Systematic errors are due all possible inaccuracies of the experimental systems used. They were at the minimum as the used experimental setup was at its best during. Errors of one data set (for example for 60 °C) in Fig. 3 are positively correlated to another data set (for 70 °C). Both bulk and track etch rates increase monotonically, deviating from the linear trend, with the concentration of the etching solutions at all temperatures.

Consider a piece of CR-39 detector containing charged particle latent tracks. The thermodynamic parameters ΔG, ΔH and ΔS are related by the following relationship,

$$\Delta H = \Delta H - T\Delta S \qquad (1)$$

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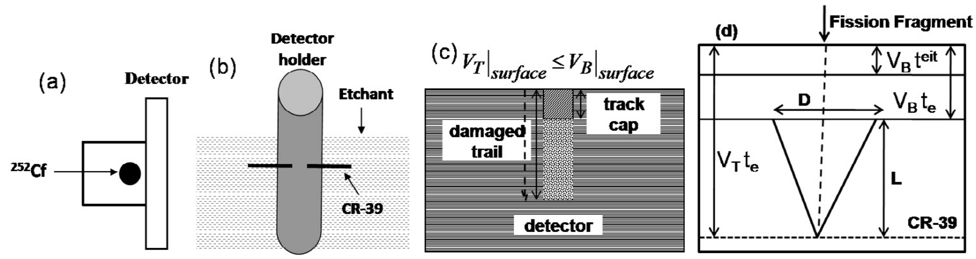

Fig. 1. Schematic of (a) exposure of CR-39 to fission fragments, (b) chemical etching procedure, (c) important sections of etched track and (d) dimensions and formulas of different parts of a typical nuclear track. In part (c), $V_T|_{surface}$ and $V_B|_{surface}$ are, respectively, the track etch rate and the bulk etch rate at the surface of the detector. In part (d), V_T & V_B are mean track & bulk etch rates, D & L are track diameter & length, t^{eit} is etch induction time, and t_e is etching time of the detector.

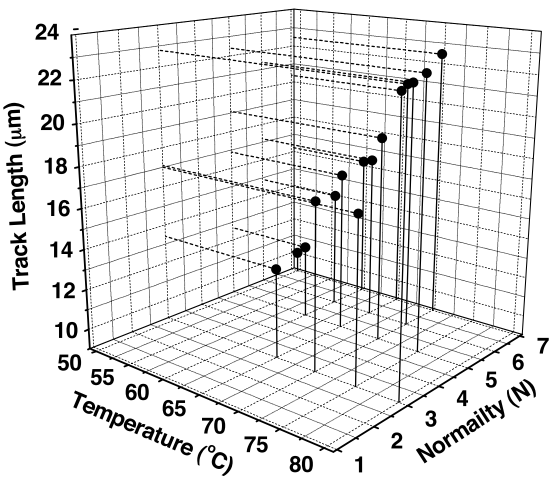

Fig. 2. Mean fission fragment track lengths observed under different etching conditions of etching solution concentration and etching temperature.

ΔG is the change in free energy, ΔH is the change in enthalpy and ΔS is the change in entropy of the sample. The part $T\Delta S$ in Eq. (1) is the increase in entropic energy of the sample. Helmholtz free energy is not a relevant parameter here as etching increases the volume of the sample. The values of were determined using the following equation,

$$\Delta G = RT[\ln(\frac{k_B T}{h}) - \ln(s/t^{eit})], \tag{2}$$

where h is Planck's constant, R is gas constant, T is the absolute etching temperature and t^{eit} is etch induction time of fission fragment tracks in CR-39. Latent tracks in a track detector do not start growing microscopically immediately after the startup of etching. Etch induction time (t^{eit}) is the time delay between the start of chemical etching of exposed detector and the time when the tracks start growing from the zero size. Details of the determination of t^{eit} were reported elsewhere [8–10]. Eq. (2) is based on a well-known formula $K^* = \frac{k_B T}{h} e^{-\frac{\Delta G}{RT}}$ [11], where K^* is the reaction rate constant.

The experimental observables (s and t^{eit}) used in Eq. (2) and determined values of ΔG are given in Table 1. Mean values of ΔG at used temperatures are plotted in Fig. 4(a). Values of free energy are averaged over etching solution concentrations in the experiments. Positive values of free energy in the plot indicate that etching process is not spontaneous at used temperatures. The increase of free energy with etching temperature in Fig. 4(a) indicates that, at higher temperatures, continuously larger amount of energy becomes available for the removal of damaged material from fission fragment trails in the target. Results in this figure show a linear dependence of free energy on the etching temperature. The linear equation with the precise values of fit parameters is,

$$\Delta G = (-0.0663 \pm 0.0066) + (0.00296 \pm 0.00025)T, \tag{3}$$

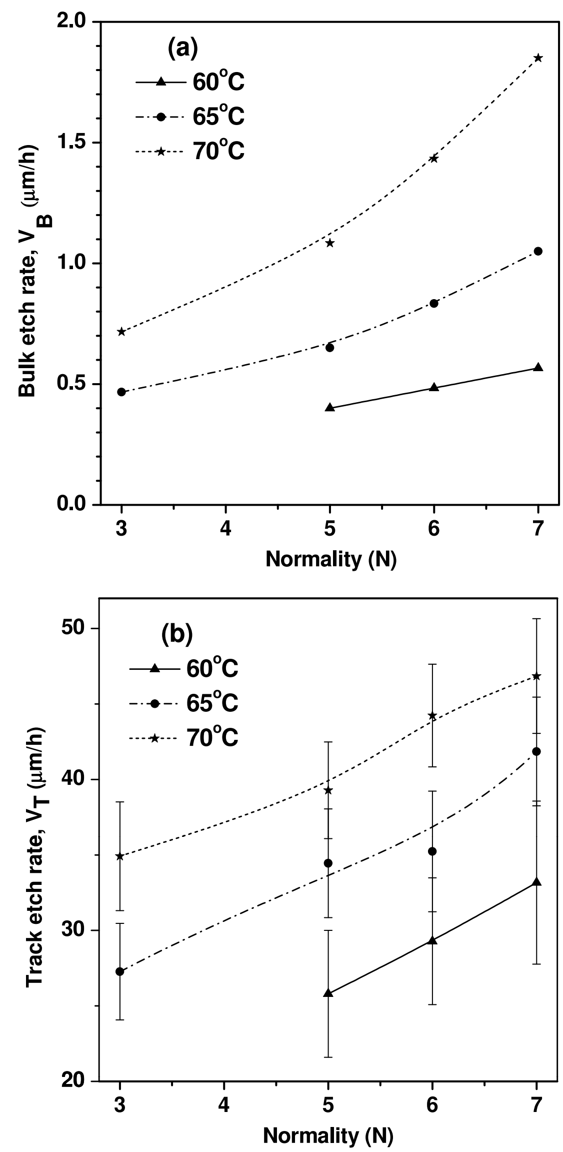

Fig. 3. (a) Bulk etch rates of CR-39 under different etching temperatures and etching solution normality or concentrations. Bulk etch rates were determined using the diametric measurements of the largest fission fragment tracks. (b) Mean track etch rates of fission fragments in CR-39, determined under the same etching conditions as above. Uncertainties in bulk etch rates are too small to be visible. Uncertainties in track etch rates are measurement. They include both systematic and statistical parts.

where T is the temperature of the etching solution in Kelvin and ΔG is the change of free energy in eV. The value of regression coefficient R