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Characteristics of the copolymerized CR-39/DAP track detector for the observation of ultra heavy nuclei in galactic cosmic rays

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ABSTRACT

A new copolymer of CR-39 and DAP resin was developed for the selective detection of ultra heavy nuclei in galactic cosmic rays. The track registration sensitivity for heavy ions with various Z^*/β was verified for copolymers mixing CR-39 (BARYOTRAK) with various concentrations of DAP using heavy ion beams at the HIMAC accelerator. In order to realize sufficient polymerization of CR-39/DAP copolymer, the optimum fabricating method was also verified by modifying the concentration of the initiator and temperature–time curing cycle for the copolymerization. CR-39 (BARYOTRAK) used for the base material of copolymerization has many advantages: 1) high uniformity of detector response, 2) extremely clear surface condition after etching, and 3) excellent charge and mass resolutions for heavy ions. The surface condition of the copolymer after etching is remarkably improved by using the BARYOTRAK monomer and its track registration sensitivity is controllable by changing the DAP concentration. However the detection threshold of the CR-39 (BARYOTRAK)/DAP copolymer is not yet satisfactory.

1. Introduction

The ability to precisely measure the chemical composition of ultra heavy (UH) nuclei ($Z > 30$) in galactic cosmic rays (GCRs) would be an important step towards resolving the remaining problems in astrophysics (Hasebe et al., 2006; Kodaira et al., 2009). Although the elemental composition of GCRs has been observed by previous experiments, the individual nuclear charge has not been sufficiently resolved. Moreover, these isotopes have not been observed due to the extremely low flux of UH nuclei in GCRs and the difficulty of charge identification of such massive particles. High performance cosmic ray detectors with excellent charge resolution and high collecting power are required to observe precisely such massive particles in GCRs. A plastic track detector such as CR-39 is one of the candidate detector for the large-scale observation of UH nuclei in GCRs, but a large number of background tracks produced by iron nuclei and other particles lighter than iron will cover up the tracks of UH nuclei. Therefore, it is important not only to raise the detection threshold of heavy ions in order to suppress background tracks produced by lighter ions with $Z/\beta < 30$ but also to achieve excellent charge resolution.

For this purpose, the performance of CR-39/DAP (diallyl phthalate) copolymer detector was verified in order to selectively detect ultra heavy nuclei (Kodaira et al., 2008a). In the previous study, it was found that track registration sensitivity and detection threshold could be controlled with the increasing the concentration of DAP. However, the surface condition after etching becomes very rough for the copolymer with high concentrations (>30%) of DAP. This indicates that the CR-39 monomer was not pure and probably contained considerable amounts of oligomers. Moreover, the CR-39 detector fabricated by the same monomer previously used for CR-39/DAP copolymer had a gradual response curve against the difference in Z^*/β. Here, Z^* and β are the effective charge of an ion and its velocity, respectively. Therefore, in order to resolve these problems in the CR-39/DAP copolymer, we exchanged the impure CR-39 monomer for a specially purified monomer of CR-39 (BARYOTRAK) manufactured by Fukuvi Chemical Industry in Japan. The advantages of BARYOTRAK as a track detector include: 1) high uniformity of detector response, 2) extremely clear surface condition after the etching, and 3) excellent charge (including mass) resolution for heavy ions (Kodaira et al., 2004, 2007).

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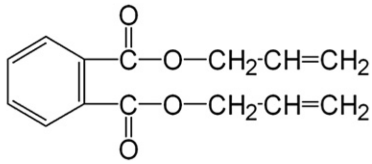

Fig. 1. Structural formula of monomer of DAP ($C_{14}H_{14}O_4$).

Table 1
Mixing rates of BARYOTRAK and DAP (wt%) for the copolymerization.

Label (%)	DAP0%	DAP10%	DAP20%	DAP30%	DAP40%	DAP50%
CR-39 (BARYOTRAK)	100	90	80	70	60	50
DAP	0	10	20	30	40	50

We also attempted to raise the detection threshold by using PEW (Potassium–Ethanol–Water), described elsewhere (Kodaira et al., 2008b).

In this paper, we summarize the performance of the CR-39 (BARYOTRAK)/DAP copolymer as a nuclear track detector and its optimum fabrication condition.

2. BARYOTRAK/DAP copolymer

Fig. 1 shows a structural formula of the DAP (ortho-type diallyl phthalate) monomer. Plastic track detectors containing a benzene ring in its structure such as DAP are empirically known to suppress track registration sensitivity for lower Z/β ions, because the energy transferred along the passage of an ion in the detector should be strongly absorbed in its benzene ring. Therefore, it is expected that track registration sensitivity for heavy ions in CR-39 detector would be suppressed by combining with DAP resin (Kodaira et al., 2008a).

DAP has three structural isomers, i.e. ortho-, iso- and para-types. Although iso- and para-type DAPs were extensively studied for use in nuclear track detectors, it was found that these types of DAP were unusable for track detection (Fujii, 1984; Schulz et al., 1991). It is known that only ortho-type DAP is able to detect heavy ions as a nuclear track detector (Tsuruta, 1999, 2000; Tsuruta et al., 2008; Koguchi and Tsuruta, 2005). In this study, we fabricated a new BARYOTRAK/DAP copolymer from the mixture of CR-39 (BARYOTRAK) and ortho-type DAP monomers.

BARYOTRAK/DAP copolymers with various DAP concentrations (see Table 1) were polymerized between two glass plates separated by a spacer of 0.9 mm thickness adopting two different temperature–time curing conditions. One is the same temperature–time curing cycle (Type A: start 45 °C, 4 h → rise linearly to 50 °C, 0.5 h → 50 °C, 3.5 h → 65 °C, 4 h → rise linearly to 80 °C, 3 h → 80 °C, 1 h) as that used for the polymerization of BARYOTRAK (Ogura et al., 1997). The curing cycle of Type A is adjusted to the copolymerization of the CR-39 detector. Therefore, there is a possibility that the polymerization of CR-39/DAP should be insufficient, because the glass transition temperature of DAP is much higher than that of CR-39.

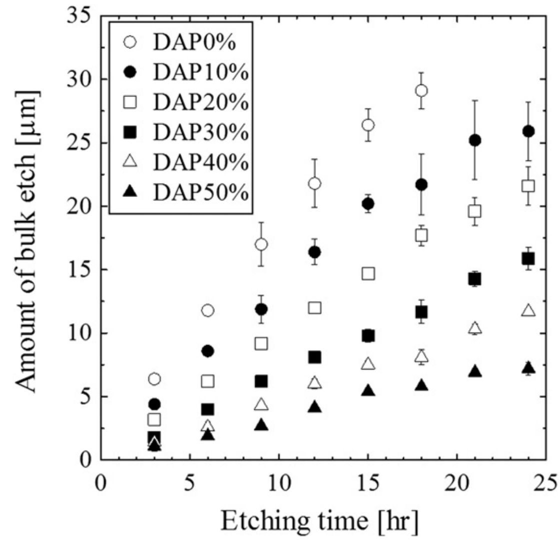

Fig. 3. Relation between etching time and the amount of bulk etch for copolymers from DAP0% to DAP50%. The linearity of V_b becomes distorted, when the fission track was over-etched beyond its residual range.

BARYOTRAK/DAP copolymers of DAP0%, 10% and 20% were also polymerized using a second temperature–time curing cycle (Type B). The curing cycle of Type B is additional heating at the temperature of 120 °C for 1 h after the curing of Type A. In order to obtain sufficient copolymerization of CR-39/DAP copolymer, the curing cycle of Type B is intended to terminate the curing process at a higher temperature than Type A. Furthermore, for the same purpose, the concentration of initiator IPP (diisopropyl peroxy dicarnonate) was varied for 3.3% (standard), 5.0% and 7.0%.

3. Experiment

BARYOTRAK/DAP copolymers were exposed to heavy ions from Si to Xe with the energies of <500 MeV/nucleon, covering the range of $19 \leq Z^*/\beta \leq 115$, in the Heavy Ion Medical Accelerator in Chiba (HIMAC) at the National Institute of Radiological Sciences (NIRS). The incident angle was normal to the detector surface. The exposed density was ~2000 ions/cm^2 for each detector. Copolymers were also exposed to the spontaneous fission fragments from ^{252}Cf to measure the amount of bulk etch and then etched in 7 mol/l sodium hydroxide solution at the temperature of 70 °C. Etching time was between 8 and 48 h depending on the range (~15 μm) of fission fragments in the detector.

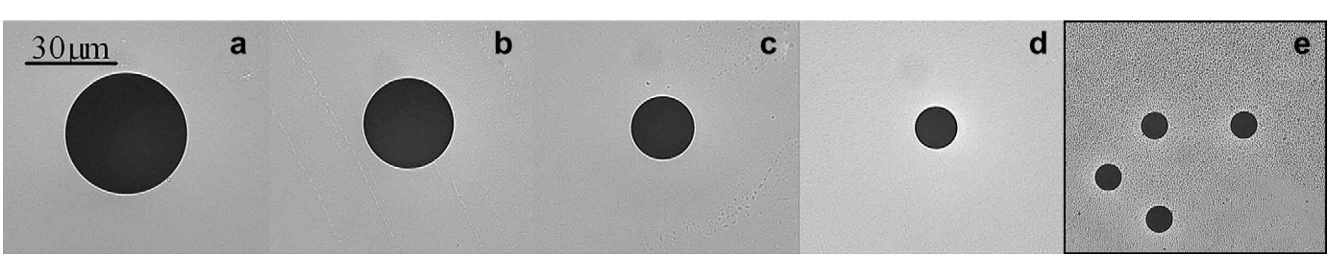

Fig. 2. Etch pit images for 189 MeV/n Xe ion. (a) DAP0%, (b) DAP10%, (c) DAP20%, and (d) DAP30%, and (e) CR-39/DAP30% used in previous work (Kodaira et al., 2008a) for the comparison of surface condition with (d) BARYOTRAK/DAP30%.