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# Thermal Neutron Monitoring Using Cellulose Nitrate Track Detector by Means of Spark Counter

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Cellulose nitrate films were examined for thermal neutron monitoring in contact with several kinds of $(n, \alpha)$ converter materials such as a plastic sheet doped with 1% boron, a solidified boron oxide plate, a single crystal plate of boron oxide and that of lithium fluoride. After thermal neutron irradiation in a research reactor, the films were etched in alkaline solution. The etch-pits on the films were counted with a spark counter. The single crystal converter of lithium fluoride was found to be the most sensitive for thermal neutron. A method for producing the single crystal converter was described. Linear relation between thermal neutron fluence and spark counts was found to be in the fluence range of $2\times10^6\sim3\times10^7 n/cm^2$ in case of using a lithium fluoride single crystal converter.

**KEYWORDS**

boron doped sheet, boron oxides, cellulose nitrate, doped materials, lithium fluorides, monocrystals, neutron fluence, neutron monitoring, spark counters, thermal neutrons, track detector.

## I INTRODUCTION

Neutron monitoring around nuclear installations is important as well as $\gamma$-ray monitoring for radiation protection. In the surrounding of reactor and accelerator, neutrons have a wide energy range from thermal to fast and are also accompanied by $\gamma$-ray. Therefore for neutron monitoring in the fields a nuclear track detector using plastic film is much more effective than a film badge or a thermoluminescence dosimeter, since the track detector is sensitive to not $\gamma$-ray but thermal and fast neutrons.

Polycarbonate and cellulose nitrate films as solid state track detectors have been used for the detection of neutrons. Perforations created by the etching of fission-fragment tracks in thin polycarbonate films can be counted automatically by an electric discharge method which is called spark counting.[(1)~(3)] This application necessitates the use of fissile materials such as $^{235}U$ or $^{232}Th$ as a converter by $(n, f)$ reaction whose widespread distribution, for example in personal dosimeters, is subject to legal and radiotoxicity resprictions. Instead thin cellulose nitrate film in contact with a lithium or boron plate as a $(n, \alpha)$ converter is also an excellent thermal neutron detector. Etch-pits produced on the film by neutrons can be counted with a spark counter.

Becker *et al.* succeeded $\alpha$-track spark counting on a self-made cellulose nitrate film in 1969.[4] He also examined spark counting of fast-neutron-induced recoil particle tracks in cellulose nitrate film (LR-115) 8 $\mu$m thick made by Kodak Pathé in France in 1975.[5] Hahn *et al.* measured thermal neutrons by the reaction $^6Li(n, \alpha)^3He$ with cellulose nitrate film made by addition of dioctyl phthalate as plasticizer in 1975.[6] We also examined spark counting of etched $\alpha$-tracks produced on a cellulose nitrate film LR-115 Type II Stripping and LR-115 Type II Stripping B (Type II Stripping coated with $LiBO_2$) in 1979.[9] Tsuruta *et al.* prepared boron doped cellulose nitrate film for spark counting of $(n, \alpha)$ etch-pits in 1982.[10]

Physical and chemical forms of converter materials such as lithium or boron influence much to the production of $\alpha$-tracks on cellulose nitrate detector. However systematic description or comparison with regard to converter forms are not found in the above and other reports up to now. Then we have prepared several kinds of lithium and boron compounds with different densities and compared their detecting efficiencies.

## II EXPERIMENTAL

### 1. Detector Film

Cellulose nitrate film "LR-115 Type II Stripping" made by Kodak Pathé Co. in France was used as a detector film. The size of this film backed with a plastic base 0.1 mm thick was 60×20 cm² and the thickness 13 $\mu$m. A sample of the film was made by cutting into the size 4×4 cm². After neutron irradiation the film was separated from base for etching.

### 2. Preparation of $(n, \alpha)$ Converters

A single crystal of lithium fluoride and several kinds of boron physico-chemical forms such as a single crystal of boron oxide, a solidified boron oxide plate, and a cellulose nitrate sheet doped with 1% boron were examined as converters.

The production of a lithium fluoride single crystal (LiF) was performed by Bridgeman method. Lithium fluoride powder in a graphite crucible heated at 900℃ in vacuum was cooled slowly from the bottom tip of the crucible. Detailed description of this procedure is shown in Ref. (11). The dimension of the single crystal was 16 mm in diameter and 27 mm in length. The single crystal thus produced was cut along the crystal axis to be formed into a plane plate as a converter.

Boron oxide single crystal converter ($B_2O_3$) was made in the same way. In this case boron oxide was heated to 630℃ (m. p.: 577℃). The lithium fluoride single crystal was easily separated from the graphite crucible, but in case of boron oxide single crystal separation from the crucible was so difficult that the crystal was taken out by breaking the graphite crucible. Cutting boron oxide single crystal along the crystal axis was also difficult. We made a boron oxide plate by filing and polishing. This diffeculty of cutting was probably due to the type of crystal. Crystal type of boron oxide is hexagonal, while that of lithium fluoride cubic.

Solidified boron oxide was made as follows: Boron oxide powder was melted at 630℃ in an oven and was cooled gradually.

The cellulose nitrate sheet containing ortho-carborane by evaporation method was prepared. The sheet contained natural boron at concentration of 1w/o.(10)

**3. Neutron Irradiation**

The films in contact with converters were irradiated with neutrons in Kinki University Reactor for 5～60 min. The maximum thermal power of the reactor was 1 W, and thermal neutron flux (fluence rate) was $2\times10^{7}\ n/cm^{2}\cdot s$ at the center of irradiation field. A film without a converter was also irradiated at the same time and place. Gold foil was also irradiated at the same time and place for measuring thermal neutron flux. In the field there were also fast neutrons, the amount of which was approximately 1/10 of thermal neutrons.

**4. Etching**

In order to find the optimum etching time, irradiated and non-irradiated films were etched with 26% NaOH solution at 50℃ for 80～250 min. Then these films were rinsed with tap water and dried. The number of etch-pits was counted with a spark counter. The relation between etching time and spark counts is shown in **Fig. 1**. (Since standard deviations of spark counts were within a few percent, the error ranges are included in the circles showing measured points in Figs. 2～4.) Net counts were derived by subtracting the counts of non-irradiated detector film from irradiated one. The left end of maximum net counts (the dotted line) corresponded to the optimum etching time 200 min. Exceeding this etching time, the curve of non-irradiated film rose abruptly.

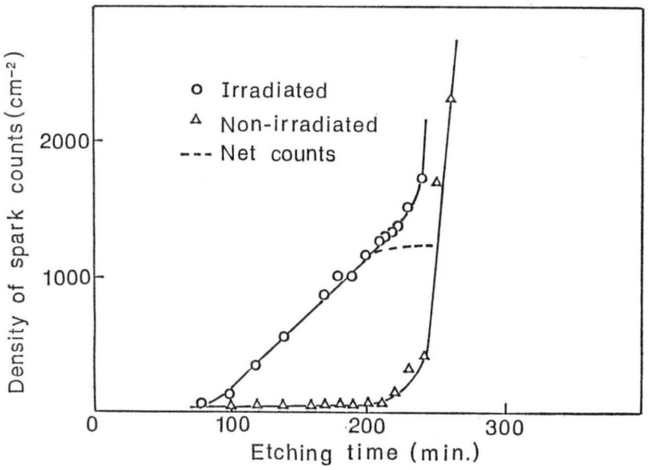


**Fig. 1** Relation between etching time and density of spark counts

**5. Spark Counting**

A cellulose nitrate film was sandwiched with a copper electrode of 22 mm diameter (anode) and an aluminized polycarbonate electrode (cathode). A high voltage (600 V) was applied between the two electrodes for punching the film. Then the aluminized polycarbonate electrode was renewed and a lower voltage (425 V) was applied again for counting.

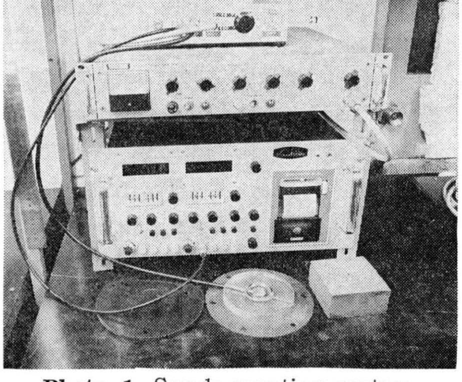

**Photo. 1** Spark counting system

Number of successive spark discharges was counted with a scaler (**Photo. 1**).

### 6. Relation between Thermal Neutron Fluence and Spark Counts

**Figure 2** shows the relation between thermal neutron fluence and spark counts for the samples using four kinds of converters. The spark counts increase in proportion to the thermal neutron fluence in one or two decades. The converters shows different detecting efficiencies by the physico-chemical form of boron. The sheet containing boron yields the minimum sensitivity. Comparing the single crystal and solidified plate of boron oxide, single crystal form was found to be more efficient converter. Spark counts of the detector film irradiated with lithium fluoride single crystal converter were several times higher than those with boron oxide single crystal converter. Therefore, single crystal of lithium fluoride was more efficient than that of boron oxide. Measurable thermal neutron flux ranges and sensitivities for the above-mentioned converters are given in **Table 1.**

**Table 1** Measuring ranges and sensitivities

| Converter | Measuring range ($n$/cm$^2$) | Sensitivity (counts/$n$) |
|---|---|---|
| LiF single crystal | $2\times10^6\sim3\times10^7$ | $4.6\times10^{-5}$ |
| $B_2O_3$ single crystal | $5\times10^6\sim1\times10^8$ | $1.4\times10^{-5}$ |
| $B_2O_3$ solidified | $2\times10^7\sim5\times10^8$ | $5.3\times10^{-6}$ |
| 1% boron sheet | $5\times10^8\sim1\times10^{10}$ | $1.9\times10^{-7}$ |

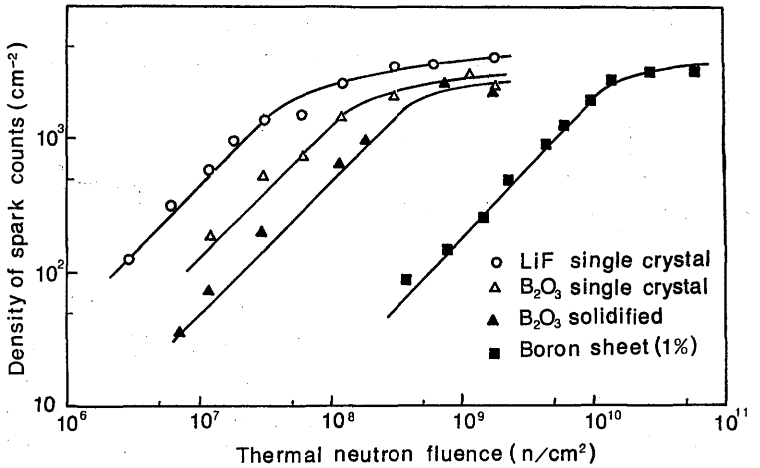


**Fig. 2** Relation between thermal neutron fluence and density of spark counts

When a cellulose nitrate detector film is irradiated with thermal neutrons mixed with fast neutrons, *e.g.* reactor neutrons, the influence of fast neutrons due to recoil charged particles should be omitted. Therefore, the spark counts of another cellulose nitrate film without converter placed with the detector film were subtracted.

## III DISCUSSION

The neutral abundance ratio of $^6Li$ and $^{10}B$ are 7.42 and 19.6%, respectively. The $(n, \alpha)$ cross section of $^6Li$ and $^{10}B$ are 945 and 3,840 b, respectively.[(12)(13)] Nevertheless, a lithium converter gave higher counting sensitivity than a boron converter. One of the reason may be considered as follows: The $(n, \alpha)$ reactions of $^{6}$Li and $^{10}$B are as follows;

$$^{6}\mathrm{Li}+{}^{1}n \rightarrow {}^{4}\mathrm{He}+{}^{3}\mathrm{H}+4.78\ \mathrm{MeV} \quad (1)$$

$$\begin{aligned} ^{10}\mathrm{B}+{}^{1}n &\rightarrow {}^{4}\mathrm{He}+{}^{7}\mathrm{Li}+2.78\ \mathrm{MeV} \quad (6.4\%) \\ &\searrow {}^{4}\mathrm{He}+{}^{7}\mathrm{Li}^{*}+2.30\ \mathrm{MeV} \quad (93.6\%) \\ &\qquad \searrow {}^{7}\mathrm{Li}+\gamma \end{aligned} \quad (2)$$

Energies of $^{4}$He and $^{3}$H in the reaction (1) are 2.05 and 2.73 MeV calculated from $Q$-value of 4.78 MeV, respectively. Energies of $^{4}$He and $^{7}$Li in the reaction (2) are 1.47 and 0.83 MeV, respectively. Alpha track forming sensitivity of cellulose nitrate detector film depends on the injecting $\alpha$-particle energy. Several kinds of thickness of polycarbonate sheets were placed between $^{210}$Po $\alpha$-source (5.3 MeV) and a cellulose nitrate detector film for a moderator. The relation between moderator thickness and counting efficiency was investigated as shown in **Fig. 3**.

The maximum sensitivity was at 20 $\mu$m thickness, which corresponded to $\alpha$-energy of 2.5 MeV. Alpha particle energy 2.05 MeV produced from lithium by the reaction (1) was closer to 2.5 MeV than $\alpha$-particle energy 1.47 MeV produced from boron by the reaction (2). Although this fact suggests the superiority of lithium fluoride single crystal over boron oxide converter, it is still insufficient as a quanititative explanation. Other major reason of higher sensitivity of lithium fluoride is not clear now. The easiness of producing lithium fluoride single crystal converter increases this superiority.

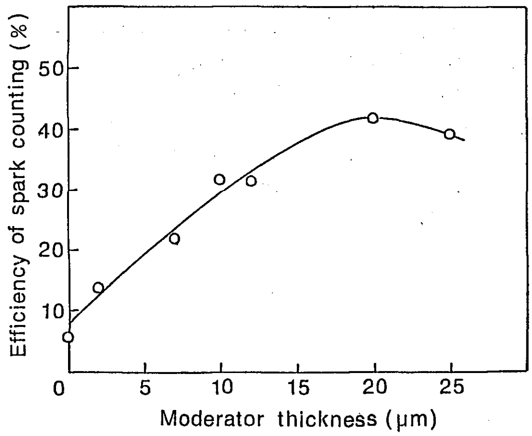


**Fig. 3** Efficiecy of spark counting as function of moderator thickness

The minimum detectable limit of thermal neutron fluence using a lithium fluoride single crystal converter is about $3\times10^{6}\,n/\mathrm{cm}^2$, which corresponds to 3 merem (0.03 mSv). (Fig. 2)

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