Jpn. J. Appl. Phys. Vol. 41 (2002) pp. 2115–2119
Part 1, No. 4A, April 2002

Neutron Emission from Fracture of Piezoelectric Materials in Deuterium Atmosphere

Masatoshi FUJII*, Hiromichi NAKAHARA, Keisuke SUEKI, Toshiaki SHIRAKAWA[1], Masami CHIBA[2], Yasushi FUJIMOTO[2], Yoshikazu HAYAMI[2], Tachishige HIROSE[2], Michiaki UTSUMI[3], Masaki HAYASHI[3], Takaharu YOKOYAMA[3] and Satoru YONEKURA[3]

Department of Chemistry, Tokyo Metropolitan University, Hachioji, Tokyo 192-0397, Japan
[1]*Department of Social Information Processing, Otsuma Women's University, Tama, Tokyo 206-8540, Japan*
[2]*Department of Physics, Tokyo Metropolitan University, Hachioji, Tokyo 192-0397, Japan*
[3]*Department of Applied Physics, Tokai University, Hiratsuka, Kanagawa 259-12, Japan*

(Received August 6, 2001; accepted for publication December 17, 2001)

We have studied nuclear reaction during a fracture process of piezoelectric materials in D_2 atmosphere. Neutrons are generated in d–d nuclear reaction through the fracture process of $LiNbO_3$ (lithium niobate) at the D_2 gas pressure of 30 kPa or above; the emission rate is 108 ± 27 /h with the statistical significance of $4.0\,\sigma$, although at 18 kPa or below, neutrons are not generated. We have not observed neutrons from other tested piezoelectric materials. [DOI: 10.1143/JJAP.41.2115]

KEYWORDS: $LiNbO_3$, piezoelectric, fracture, mechano-reaction, acceleration, deuterium, pressure, neutron

1. Introduction

It has been reported that a mechanical effect on deuterated metals and piezoelectric materials gives rise to d–d nuclear reaction.[1–6] In this reaction a 2.45 MeV neutron is generated with a ^{3}He nucleus. In a crushing process of deuterated metals in deuterium gas, neutron emission from d–d nuclear reaction has been reported by Lipson and co-workers,[1,3] Deryagin *et al.*[2] and Klyuev *et al.*[4] For this reaction, they utilized materials with high deuterium contents.

In contrast, we employed piezoelectric materials, which generated a high electric field when a high strain force was applied. From the results of previous experiments, Shirakawa *et al.*[5] reported that neutrons were generated in a fracture process of $LiNbO_3$ (lithium niobate) single crystal in deuterium gas at 101 kPa, while we did not observe neutrons in hydrogen gas at 101 kPa.[6] This phenomenon was called mechano-nuclear reaction.

In this paper, we report the dependence of neutron emission rate on deuterium gas pressure in the fracture process of $LiNbO_3$ single crystal in order to confirm the previous experimental results.[5,6] Other piezoelectric materials are also surveyed to determine whether they generate neutrons at 101 kPa.

2. Experimental Procedure

The materials were fractured by a vibro-mill (VP-100, ITOH Co., Ltd., Japan) with a gas-tight stainless-steel, SUS304, cell. The cell had a hemispherical inner shape with a radius of 5 cm, and the height from the ceiling to the bottom was 6 cm. The total inner volume was 230 cm^3. A steel ball with the diameter of 5 cm (514 g) was put together with 12 g of the material. After evacuation of the cell, deuterium gas was injected at a prescribed pressure between 0.036 and 1000 kPa. The cell was vibrated at 50 Hz with an amplitude of 0.5 cm up and down, so that the steel ball moved up and down in the cell and hit the ceiling. The crystal was crushed to powder (about 1 μm in diameter) after 1 h of vibration, but the diameter did not decrease with further vibration. Thus, each fracture experiment was stopped at 1 h. Then both the sample and the gas were exchanged with new materials and the fracture experiment was repeated. We carried out five fracture experiments in one day.

The neutrons were detected by ten ^{3}He proportional counters (Reuter-Stokes RS-P4-0806-207), with the diameter of 2.54 cm and the effective length of 15.09 cm, pressurized at 4 atm. They were inserted circularly in a full barrel-shaped paraffin block of 47 cm outer diameter and 19 cm inner diameter with the height of 25 cm (Fig. 1). The paraffin block was used for thermalizing the neutrons to increase the detection efficiency. The cell was set at the center of the paraffin block. The ten ^{3}He counters were divided into five sets of two counters. The pulse height of each set was digitized by analog-to-digital converters (12-bit ADC) in the range between 0 and 4095 channels independently. Signal processing and data recording were carried out by standard nuclear instrument modules (NIM) and computer automated measurement and control (CAMAC) systems.[7] When the sum of the pulse height in the five sets exceeded channel 600, the event was recorded along with the pulse height, the scalar count of each counter set and the event time. Usually only one of the five sets exceeded channel 600.

The pulse height was calibrated by a ^{252}Cf neutron source, as shown in the Fig. 2 inset. Total energy deposited to the ^{3}He gas counter by the thermalized neutrons is 760 keV.

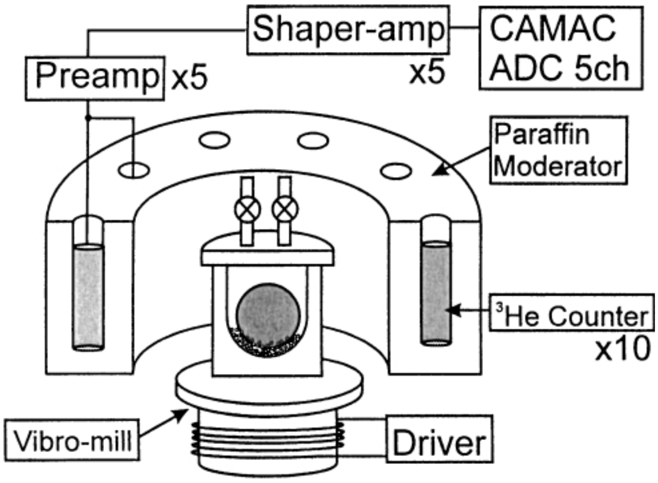

Fig. 1. Schematic diagram of the apparatus.

*E-mail address: fujii-masatoshi@c.metro-u.ac.jp

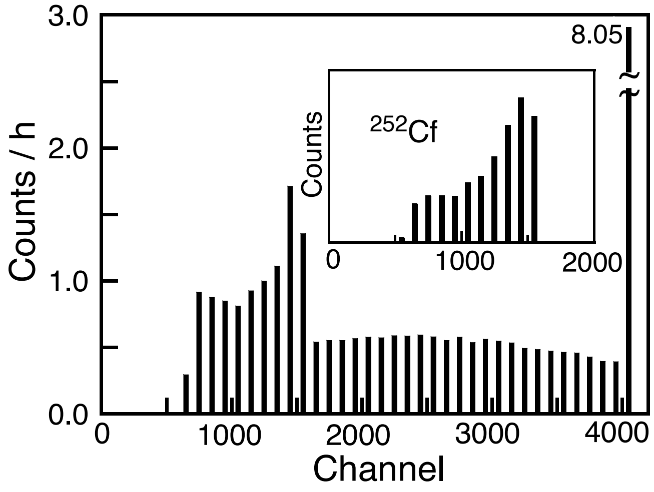

Fig. 2. Pulse height distribution of the BG counts/h accumulated for 2880 h. Inset shows that of ^{252}Cf source for 3 min. The peak position is set at 1500 channel of the ADC.

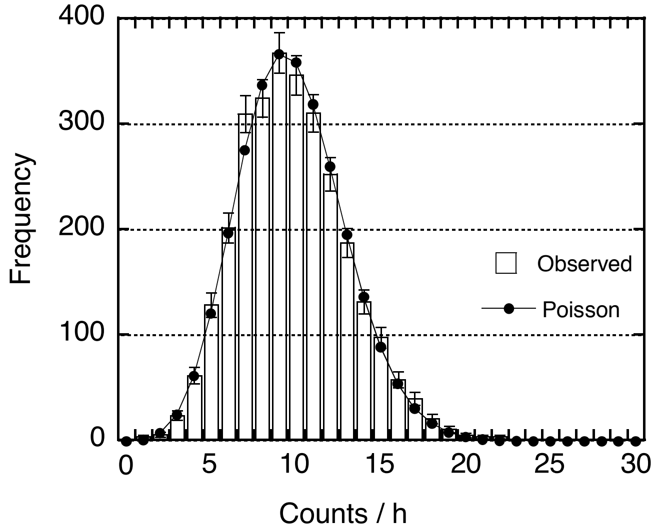

Fig. 3. Frequency distribution of the BG counts/h for 2880 h. Average is 9.78 ± 0.06 counts/h. Bars are observed frequency and dots are Poisson distribution of the mean value at 9.78. From chi-squared test ($\chi^2/\nu = 10.9/17$), the confidence level of the consistency is 0.861.

Each counter was tuned to the total energy deposit at channel 1500 of the ADC by the high-voltage power supply to the counter. The partial energy deposit signals were distributed in the lower channels. Zero energy deposit was at channel 60. Thus, we selected the signal between 600 and 1599 channels to be considered as that of a neutron.

The experiment was carried out in the low background (BG) facility at Nokogiri Mountain of The Institute for Cosmic Ray Research, The University of Tokyo (ICRR). The facility was located underground to reduce BG due to cosmic rays.[8)] To prevent the generation of electric noise induced by electric leakage at the high-voltage connectors of the counters, we blew dry air on the counters and maintained dry conditions of less than 50% RH. To evaluate the environmental conditions, we monitored and automatically recorded the humidity, the temperature, and the atmospheric pressure at the counters and the electronics every hour. Background neutrons were measured under the same experimental conditions as the fracture processes without vibration: the cell contained a fractured sample and the prescribed pressure of deuterium gas. To reduce the statistical error and to confirm the deviation from the statistics, we measured the BG for as long as possible;[9)] the BG measurements were continued during the fracture experimental period of four months, except while the series of five fracture experiments was proceeding.

Figure 2 shows the pulse height distribution of the BG counts/h accumulated for 2880 h, which is composed of neutron signals and flat counter noise. The shape and the peak position of the neutron signal coincide with those of the thermalized neutron from ^{252}Cf (inset of Fig. 2). In order to reduce the BG, events were discarded by a software cut when the signals of two or more sets of the counters exceeded the pulse height of 600 channel within 3 μs, or the signals of all sets exceeded channel 100. The detection efficiency of a neutron was measured to be 2.6% by the calibrated ^{252}Cf source set at the center of the cell. The detection efficiency determined by the calibrated ^{252}Cf source was verified by a Monte-Carlo simulation of the neutron transport code of MCNP.[10)] Two cases were studied with respect to the energy distribution of neutrons emitted from the source. For the first case, one million monochromatic neutrons of 2.45 MeV were generated at the center of the cell with a steel ball; the paraffin moderator and the ^{3}He counters were set as in the experiment. Energy was expected to be produced in the $d + d \rightarrow n + {}^3He$ reaction. For the second case, one million neutrons were generated with the fission energy distribution from the ^{252}Cf source at the center of the paraffin moderator. The energy distribution of neutrons arriving at the ^{3}He counters were recorded and compared for both cases. Less than 0.3 eV neutron energy, the shapes of energy spectra as well as the number of neutrons entering the ^{3}He counters coincided within 1%. The main contribution producing the pulses in the ^{3}He counters is $n + {}^3He \rightarrow p + t$ around the thermal energy of neutrons less than 0.3 eV. Therefore, we concluded that the detection efficiencies of neutrons from the $d + d$ reactions and ^{252}Cf source were the same.

The total BG neutron count with the software cut was 28178 counts for 2880 h, 9.78 ± 0.06 counts/h.

The frequency distribution of the neutron counts/h and a Poisson distribution of the mean value at 9.78 are shown in Fig. 3. The frequency distribution was consistent with the Poisson distribution and the confidence level of the consistency was 86% by chi-squared test. Therefore, the BG events took place independently during the experimental fracture period. The BG neutron counts immediately before and after each series of fracture experiments were also consistent with 9.78 ± 0.06 counts/h. Thus, we can take the value of 9.78 ± 0.06 counts/h as the BG value for the following statistical analysis of the fracture experiments.

3. Results and Discussion

We measured the neutron emission rates utilizing $LiNbO_3$ single crystal with respect to the deuterium gas pressure from 0.036 to 1000 kPa in the fracture process. The crystal was the top part of a 3 inch Czochralski single crystal rod having the ⟨0114⟩ growth direction, manufactured by

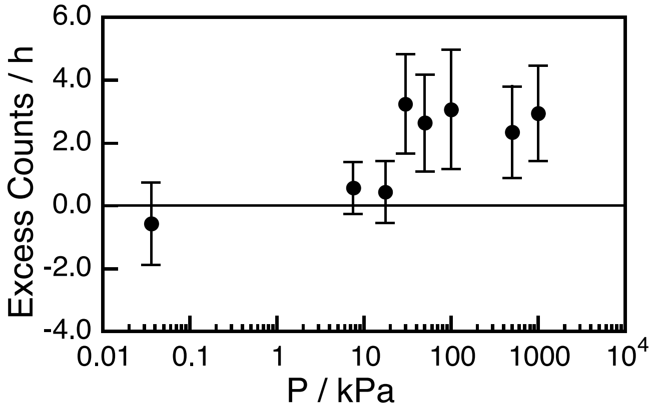

Fig. 4. Excess neutron counts/h calculated by subtracting the BG counts from the counts during the fracture process of $LiNbO_3$ from 0.036 to 1000 kPa in deuterium gas. The data at 101 kPa are taken from our previous results.[5)]

Toshiba Co.[11)] The Curie temperature measured by the DTA method was 1140°C. Thus, the composition of Li_2O was 48.4 mol% [$Li_2O/(Li_2O + Nb_2O_5)$]. Figure 4 shows excess neutron counts with one standard deviation, which were calculated[9,12)] by subtracting the BG counts from the measured counts in the fracture process of $LiNbO_3$ for eight different pressure conditions of deuterium gas. The excess neutron counts at 101 kPa are taken from our previous results,[5)] where the experimental conditions were the same as in this experiment.

The excess neutron counts increase stepwise with increasing pressure. In the case of deuterium gas at a pressure not more than 18 kPa, the excess neutrons are not measured. The excess neutron counts increase at 30 kPa and saturate. Therefore, we divide the data into two pressure regions, 18 kPa or below called the lower pressure region and 30 kPa or above called the upper pressure region. The weighted mean value between 30 kPa and 1000 kPa is 2.82 ± 0.70 counts/h. The neutron emission phenomenon that was observed in the previous experiment[5,6)] is reproduced with the statistical significance of $4.0\,\sigma$. Then we carry out a statistical analysis of the excess neutron rates for each region. The results given in Table I show the weighted mean values (μ), reduced chi-square values (χ^2/ν) for a null generation hypothesis (ν is the number of degrees of freedom), and confidence level (CL) of the neutron generation for each pressure region.

If the excess neutron rates in both the lower and the upper pressure regions are assumed to be constant and consistent with no neutron generation, 1-CL values are 64.9% and 0.3%, respectively. Thus, CL of neutron generation becomes 99.7% in the upper pressure region. If the excess neutron rates in the upper pressure region are assumed to be constant at 2.82 counts/h, which is the weighted mean value of excess neutron counts (μ) in the upper pressure range, the reduced chi-square value ($\chi^2/4$) and CL are 0.05 and 99.5%, respectively.

From these results, it is claimed that excess neutrons emitted in the fracture process at 30 kPa or above and the emission rates are constant at 108 ± 27 emissions/h, which is calculated from the count rate of 2.82 ± 0.70 counts/h and the efficiency of 2.6%, regardless of the pressure. In order to verify whether residual noise events remain after subtraction of the BG, we analyze the pulse height distribution for each pressure region.

Figure 5 shows the pulse height distributions without the software cut, which are derived by subtracting the BG pulse height distribution from the pulse height distribution in each pressure region, normalized by one hour. The distribution in the upper region, Fig. 5(a), resembles that of the ^{252}Cf source, shown in Fig. 2. The characteristic distribution ranges from 600 to 1599 channels, where the sum of the excess counts, 2.79 counts/h, corresponds to the weighted mean value of the excess neutron rates of 2.82 ± 0.70 counts/h with the software cut. In contrast, above channel 1599, the sum of the excess counts is -0.04 counts/h. On the other hand, for the lower pressure region, there is no apparent distribution feature for the entire range of channels, Fig. 5(b). In fact, the total excess count from 600 to 1599 channels is -0.14 counts/h and that above channel 1599 is 0.19 counts/h. Thus, the systematic error for the neutron count is estimated to be no more than $|-0.14|$ counts/h. Furthermore, for the upper pressure region the total number of excess counts is estimated to be zero, other than the counts between 600 and 1599 channels. Therefore, the observed excess counts/h 2.82 ± 0.70 is identified as the number of detected neutrons from the fracture process.

Table I. Weighted mean values of the excess neutron rates (μ), reduced χ^2 values for a null neutron generation hypothesis (χ^2/ν), and corresponding null (1-CL) and positive confidence levels (CL) for the pressure ranges of 18 kPa or below and 30 kPa or above.

P (kPa)	μ (counts/h)	χ^2/ν for null	1-CL (%)	CL (%)
≤18	0.31 ± 0.57	0.43	64.9	35.1
30≤	2.82 ± 0.70	4.05	0.3	99.7

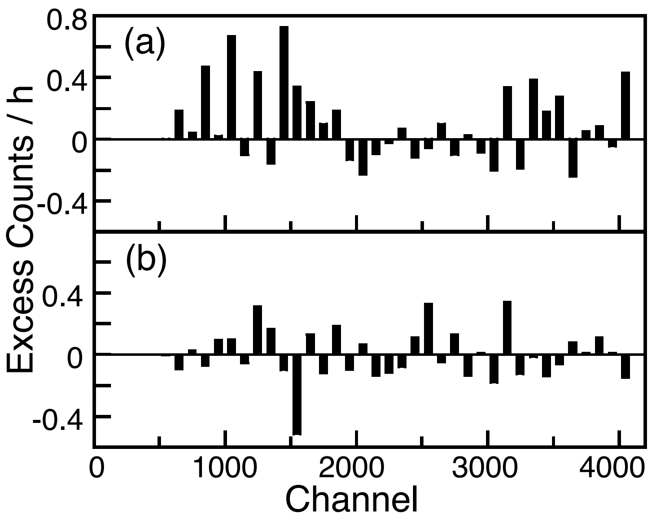

Fig. 5. Pulse height distribution of counter signals without the software cut calculated by subtracting BG signals from the counts of fracture experiments of $LiNbO_3$: (a) the sum of pulse height distribution of the upper pressure region at 30 kPa or above, and (b) that of the lower pressure region at 18 kPa or below.

Table II. List of tested piezoelectric materials with piezoelectric constant, d, dielectric constant, ϵ, and open-circuit voltage, $g(d/\epsilon\epsilon_0)$. The last column is the rate of generated neutrons, R, at 101 kPa in deuterium gas in the fracture process.

Material	d (10^{-12} C/N)	ϵ (Vm/N)	g ($d/\epsilon\epsilon_0$)	R (emissions/h)
$LiNbO_3$	69.2[a]	85.2[a]	0.092	108 ± 27[†]
$LiTaO_3$	26.4[a]	53.6[a]	0.056	32 ± 54
Rock crystal (SiO_2 crystal)	2.25[b]	4.59[b]	0.055	-6 ± 37
Tourmaline	−2.16[b]	7.51[b]	0.033	-6 ± 50
$PbZrO_3$ $+PbTiO_3$ (PZT)	320[c]	1350[c]	0.028	-56 ± 44
Rochelle salt (deuterated)	435[b]	501[b]	0.098	-61 ± 65

a) *Landolt-Börnstein* (Springer-Verlag, Berlin, 1981) III/16a.
b) *American Institute of Physics Handbook* (McGraw-Hill, New York, 1982) 3rd ed., p. 3-122.
c) Catalog data of TDK.
† The weighted mean value between 30 kPa and 1000 kPa.

In addition to the experiment of 1 h, we performed fracture experiments in D_2 and H_2 atmosphere at 101 kPa with long durations of 367 h and 611 h, respectively. In the D_2 experiment, the neutron count rate was 10.10 ± 0.17 counts/h, which was slightly higher than the BG rate (9.78 ± 0.06 counts/h), but lower than the original experiment. This means that the neutron emission does not continue for a long time. In the H_2 experiment, the neutron count rate was 9.93 ± 0.13 counts/h, which was consistent with the BG rate. This means that the neutron counters do not sense noise induced by the vibration and discharge in the cell.

The mechanism of this reaction is assumed to be as follows. The deuterium is ionized by an exoelectron or other charged particle in the fracture process.[13,14] Then the ionized deuterium is accelerated by the generated high electric field through a crack in the $LiNbO_3$ crystal, which is produced by the crushing process. Immediately after the creation, the space in the crack is kept in vacuum and the ionized deuterium can be accelerated. Then the accelerated deuterium hits an atmospheric deuterium with high velocity, producing sufficient d–d nuclear reaction to explain the observed neutron rates. The pressure dependence of the neutron generation, which resembles a step function, indicates that the reaction rate is not proportional to the density of the deuteron; there are some key factors other than the density.

We searched for piezoelectric materials other than $LiNbO_3$ that give rise to the mechano-nuclear reaction. When a piezoelectric material is stressed at the same strain force, the generated voltage is proportional to an open-circuit voltage, g, due to piezoelectric effect. The g value is one of the characteristic parameters of piezoelectric materials and it is defined as the ratio of the piezoelectric constant, d, to the dielectric constant, $\epsilon\epsilon_0$. For testing the mechano-nuclear reaction, we selected piezoelectric materials having high g values listed in Table II.

Single crystals of $LiNbO_3$ and $LiTaO_3$ were manufactured by Toshiba. They have the same crystal structure. Natural rock crystal (single crystal of SiO_2) and tourmaline were produced at Santade in Colombia and in India, respectively. PZT91C ($PbZrO_3 + PbTiO_3$) was manufactured by TDK. In addition to these crystals commonly used as piezoelectric materials, we employed Rochelle salt that was purchased from Wako Pure Chemical Industries, Ltd. and deuterated by us.

The last column of Table II presents generated neutron rates (R) in the cell for each material at 101 kPa, except $LiNbO_3$, in deuterium gas, accompanied by the statistical errors. R for $LiNbO_3$ is the weighted mean value between 30 kPa and 1000 kPa. The generated neutron rates in the cell are calculated by subtracting the BG counts from the measured neutron counts, then corrected by the detection efficiency of 2.6%. Among the tested materials, $LiNbO_3$ alone shows the generation of neutrons ($4.0\,\sigma$) in the cell.

Lithium niobate has one of the highest g values of the single crystals, thus high voltage is generated with this material. Having higher g values than that of $LiNbO_3$, deuterated Rochelle salt shows no evidence of neutron generation within statistical errors. Rochelle salt is polycrystalline, while $LiNbO_3$ used in our study is single crystal. Moreover, Rochelle salt is too soft to withstand for any strain force. These properties may cause the difference in the efficiency of the mechano-nuclear reaction.

In conclusion, we find that the neutrons are generated in the fracture process of $LiNbO_3$ not only at 101 kPa but also at 30 kPa or above in deuterium atmosphere. The generation rate is 108 ± 27 emissions/h ($4.0\,\sigma$) for the pressure range between 30 kPa and 1000 kPa. The confidence level by the chi-squared test is 99.5% for the estimated counting rate of 2.82 ± 0.70 counts/h above 30 kPa. At 18 kPa or below, neutron generation is not detected. Neurons are generated in the fracture process of $LiNbO_3$, but the other five tested piezoelectric materials do not show any effect of the mechano-nuclear reaction at 101 kPa in deuterium atmosphere. Although neutron generation rates do not increase up to 1000 kPa for the $LiNbO_3$ fracture process, there is a possibility that it is enhanced at the higher pressure. Carrying out the experiment at higher pressure is necessary.

Acknowledgment

We would like to express our appreciation to the late Professor K. Yamakoshi (ICRR) for his valuable support.

1) A. G. Lipson, V. N. Mordovin, B. V. Deryagin and Yu. P. Toporov: Phys. Lett. A **166** (1992) 43.
2) B. V. Deryagin, A. G. Lipson, V. A. Kluev, D. M. Sakov and Yu. P. Toporov: Nature **341** (1989) 492.
3) A. G. Lipson, D. M. Sakov, V. A. Klyuev and B. V. Deryagin: JETP Lett. **49** (1989) 675.
4) V. A. Klyuev, A. G. Lipson, Yu. P. Toporov, B. V. Deryagin, V. I. Lushchikov, A. V. Strelkov and E. P. Shabalin: Sov. Tech. Phys. Lett. **12** (1986) 551.
5) T. Shirakawa, M. Chiba, M. Fujii, K. Sueki, S. Miyamoto, Y. Nakamitsu, H. Toriumi, T. Uehara, H. Miura, T. Watanabe, K. Fukushima, T. Hirose, T. Seimiya and H. Nakahara: Chem. Lett. (1993) 897.
6) M. Chiba, T. Shirakawa, M. Fujii, T. Ikebe, S. Yamaoka, K. Sueki, H. Nakahara and T. Hirose: Nuovo Cimento A **108** (1995) 1277.
7) Y. Nakamitsu, M. Chiba, K. Fukushima, T. Hirose, K. Kubo, M. Fujii, H. Nakahara, T. Seimiya, K. Sueki, M. Katada, N. Baba, S. Kamasaki, S. Ikuta, K. Endo and T. Shirakawa: Nuovo Cimento A **107** (1994) 117.
8) T. Shibata, M. Imamura, S. Shibata, Y. Uwamino, T. Ohkubo, S. Satoh, K. Yamakoshi, N. Oyama, T. Ohsaka, N. Yamamoto, O. Hatozaki and N. Niimura: Nucl. Instrum. Methods Phys. Res. A **316** (1992) 337.
9) W. J. Price: *Nuclear Radiation Detection* (McGraw-Hill, New York, 1964) Chap. 3.
10) MCNP—A General Monte Carlo N-Particle Transport Code, Los Alamos National Laboratory Report LA-12625-M, ed. J. F. Briesmeister (March, 1997).
11) K. Yamada, H. Takemura, Y. Inoue, T. Omi and S. Matsumura: *Proc. 6th Meet. Ferroelectric Materials and Their Applications, Kyoto 1987*, Jpn. J. Appl. Phys. **26** (1987) Suppl. 26-2, p. 219.
12) A. H. Jaffey: Nucleonics **18** (1960) 180.
13) K. Nakayama, N. Suzuki and H. Hashimoto: J. Phys. D **25** (1992) 303.
14) J. T. Dickinson, L. C. Jensen, S. C. Langford, R. R. Ryan and E. Garcia: J. Mater. Res. **5** (1990) 109.