



Measurement of diffusion coefficients in solids by the short-lived radioactive beam of ^8Li

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Abstract

As an effort to effectively use the short-lived radioactive ion beams to be available in near future at Tokai Radioactive Ion Accelerator Complex, we have developed a radiotracer method for diffusion studies in solids. A test experiment has been performed to measure the diffusion coefficients of Li in the sample of LiAl compound, by using the α -emitting radiotracer of ^8Li ($T_{1/2} = 0.84$ s). The yields of α -particles emitted from diffusing ^8Li primarily implanted in the sample have been measured as a function of time under a recurrent time sequence; the implantation of ^8Li for 1.5 s (beam-on) and subsequent diffusion for 4.5 s (beam-off). We have found that the time-dependent yields of the α -particles could be used as a measure of the diffusivity of the tracer in a non-destructive way. By taking into account the experimental conditions, we have simulated the time-dependent α -particle yields for various diffusion coefficients and compared with the experimental ones in order to extract the diffusion coefficients of Li in the sample. The process of the simulations and the comparisons is discussed.

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1. Introduction

At Tandem Accelerator Facility of JAERI-Tokai, a radioactive ion beam facility, TRIAC-Tokai Radioactive Ion Accelerator Complex – is under construction [1]. In the facility, short-lived radioactive ions, produced by proton or heavy ion induced nuclear reactions, can be accelerated up to the energy necessary for experiments. The energy is variable in the range from 0.1 to 1.1 MeV/amu, allowing one to implant the radioactive ions into specimens at a proper depth. Various accelerated radioactive ion beams with intensities of 10^4 – 10^7 ions/s depending on nuclear species [2], will be available from the end of 2004 for experiments in nuclear physics, nuclear astrophysics and materials science.

As an effort to effectively use the short-lived radioactive ion beams for materials science, we have developed a radiotracer method for diffusion studies in solids. In our previous reports [3], we have simulated the feasibility of the radiotracer method for diffusion studies using short-lived radioactive ion beams as tracers. Here, the solid material of interest is set at a given temperature and recurrently irradiated (implanted) by the energetic tracer beam for a time-duration, order of half-life time of the tracer. Especially when using the α -emitting radiotracer of ^8Li (half-life: 0.84 s), the simulation has shown that the tracer diffusion coefficients in the solid can be measured in a none-destructive and on-line way, completely different from the conventional radiotracer method in conjunction with a serial sectioning technique [4]. The time-dependent yields of α -particles from the diffusing ^8Li have been examined as a direct measure of the diffusion of ^8Li in the solid material of interest, reflecting the temporal evolution of the concentration-depth profile of the ^8Li primarily implanted in the material.

For the experimental confirmation of the method, an experiment has been performed to measure

the diffusion coefficients of Li in the sample of LiAl compound, by using the α -emitting radiotracer of ^8Li . Using the recoil mass separator (RMS) of JAERI [5], the ^8Li beam was produced for the experiment. In the following, we will present and discuss the results of the test experiment.

2. Experiments

The experimental set-up for the diffusion measurement is shown in Fig. 1, installed nearly at the focal position of JAERI-RMS. The radioisotope of ^8Li was produced, by bombarding ^7Li of 24 MeV on ^9Be target of 42 μm in thickness. Separated by JAERI-RMS (not shown here), the ^8Li beam of 14.6 MeV with about 0.6 MeV in a full width at half maximum (FWHM) was obtained with an intensity of 2500 particles/s by 30 enA of ^7Li . The energy of ^8Li for implantation was further reduced to 4 MeV by an energy degrader (Al foil) installed upstream, corresponding to the average implantation-depth of about 10 μm from the front

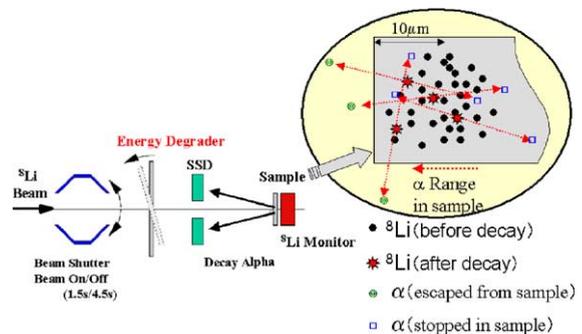


Fig. 1. Schematic, experimental set-up for the measurement of ^8Li diffusion coefficients in LiAl. The principle of the measurement is also schematically presented: The yield of α particles measured at a time is a measure of the diffused distribution of ^8Li primarily implanted in the sample with a depth of about 10 μm .

surface of the sample. One time sequence for the measurement consisted of the time-duration of 1.5 s for implantation (beam-on) and of 4.5 s for subsequent diffusion (beam-off). The beam-on and off operation was carried out by the beam shutter as shown in Fig. 1. The sample was set at 20 °C (room temperature) before starting the measurement. It should be noted that the diffusion time mentioned above is different from that of the conventional method because the tracer in the present method diffuses all the time of the measurement. This is the reason why we call the present method as an on-line measurement of diffusion. As schematically shown in Fig. 1, the implanted ^8Li decays into two α -particles, whose average range in LiAl is 8 μm . Then a charged particle detector, located close to the sample surface, could selectively detect α -particles from ^8Li diffusing toward the sample surface, since the implantation-depth is deeper than the range of α -particles in present case. Therefore the time-dependent yields of α -particles with measurable energies are supposed to be sensitive to the Li diffusion in the sample. The α particles coming out of the sample were measured as a function of time by an annular solid-state detector (SSD). The sequence was repeated to obtain good statistics, where the time-zero was always at the beginning of the irradiation. We performed the measurement in the same way for the temperature of 150 °C and 300 °C, respectively.

3. Results and discussion

The time spectrum of the yield of α -particles with measured energies larger than 400 keV is shown in Fig. 2. Also shown is the time spectrum of the yield of ^8Li alive in the sample, corresponding to the trivial time-dependence of the α -radioactivity. It gives a reference time spectrum in estimating the diffusion coefficient from the time spectra of the yield of α particles emitted by diffusing ^8Li in the sample. Once the time spectrum of the yield of alpha particles is normalized by the reference spectrum, the time spectrum associated with the temporal evolution of the profiles of diffusing ^8Li under a certain diffusion coefficient in the sample could be obtained, which is also pre-

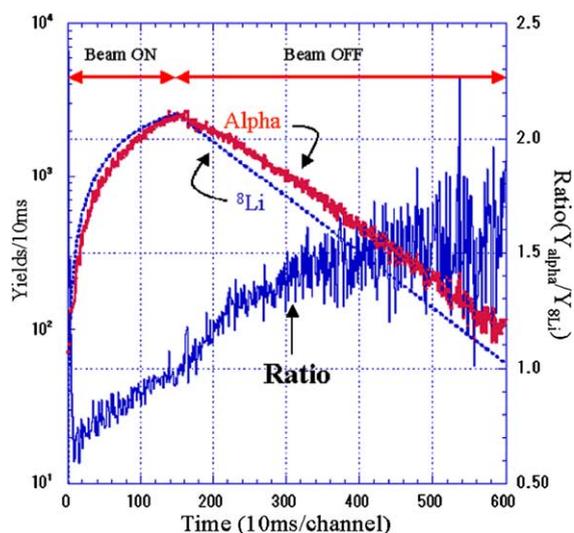


Fig. 2. Time-dependent yield spectra of α -particles and ^8Li in LiAl. The ratios, i.e. yields of α particles normalized by those of ^8Li at a given time, are also presented as a function of time (right-side scale). The yields of ^8Li are calculated according to the radioactivity of ^8Li during the measuring time-cycle and adjusted to the value of α yields at 1.5 s. The time zero is always set by the starting time of beam irradiation.

sented in Fig. 2. The normalized time spectrum is represented by the ratio, i.e. yield of α -particles divided by that of ^8Li existing in the sample at the time of interest. If ^8Li does not diffuse at all, the normalized spectrum should be constant over time. However, the experimental values of the ratios gradually increase with time, well demonstrating that more α -particles are coming out of the sample with increasing time as a result of the diffusion of the α -emitting ^8Li , otherwise stopped in the sample and thus not detected.

In Fig. 3 are shown the normalized time spectra of α -particle yields that were measured at different temperatures, 20 °C, 150 °C and 300 °C, respectively. By comparing an experimental normalized time spectrum obtained at a certain temperature, with those simulated with different diffusion coefficients, a diffusion coefficient at the temperature could be finally obtained. In Fig. 3 are also presented the time spectra simulated with the diffusion coefficients best reproducing the experimental data at the respective temperatures as a result of the comparisons.

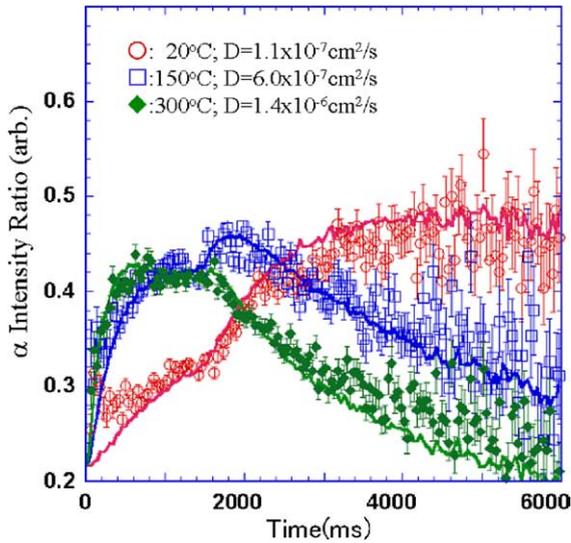


Fig. 3. Normalized time spectra of α yields from LiAl measured at the temperatures as indicated. The results from the simulation are also shown with the diffusion coefficients, which are assumed in the simulation by taking account the experimental conditions.

We here briefly describe the simulation and the process of the comparisons between the simulated and experimental spectra in order to extract diffusion coefficients. In the simulation, as described in detail in [3], we first defined the incident energy and energy spread of the ^8Li beam from the energy spectrum measured in the instant of implantation. Using the incident condition of the beam, we simulated the concentration-depth profile of ^8Li implanted in the sample and the time-evolution of the profile when a certain diffusion coefficient of Li in the sample was assumed. And then, as a measure of Li diffusion in the sample, the energy spectra of α particles emitted from the time-dependent profiles of ^8Li was simulated, by referring to the energy loss and straggling on their passage from the emitted position to the sample surface. Finally, the time-dependent α -particle yields depending on the diffusion coefficients assumed in the simulation were obtained and then compared with the experimental time spectra shown in Fig. 3, after being normalized in the same way as preformed for the experimental data. The parameters (mean and FWHM) describing the concentration-depth profile and the energy loss and straggling of α -par-

ticles were estimated by using the SRIM-2003 code [6], which is widely used in this kind of application with high reliability.

Simply guided by eye in the present comparisons, we adopted the diffusion coefficients assumed in the simulation that best reproduced the experimental time-dependence. The absolute scales of the ordinate in Fig. 3 are arbitrary, but relatively correct in different measurements. Once we adjusted parameters in the simulations in order to reproduce the data at a temperature, e.g. 150 °C in the present case, what remains for further comparisons with the data measured at different temperatures is only the variation of the diffusion coefficients accordingly. The parameters for the first fine adjustment include the absolute scale, the implantation-depth and the width of the primary concentration-profile of the tracer and the energy threshold of α -particles. Among them, the implantation-depth was found to be the most sensitive parameter and we could not reproduce the experimental time-dependence if we used the depth estimated by the SRIM-2003 code [6]. The resultant depth is shorter than the estimated value by about 5% in the target matrix of Al (energy degrader) and LiAl at the incident energy of 14.6 MeV and by about 17% in LiAl at 4 MeV. This is another interesting aspect of the measurement; the present method could be applied as a range meter of Li isotopes in the compound where the Li diffusion coefficients are well known. On the other hand, it is a disadvantage of the present method because an additional, not well-defined, parameter should be taken into account in the simulation in order to extract unknown diffusion coefficients. Consequently, the accuracy of the diffusion coefficients finally obtained in the way mentioned above is reduced to a large extent. Although more deliberated analysis for estimating the accuracy is necessary, we can conclude preliminary that the diffusion coefficients could be extracted with an accuracy of 25%, especially at room temperature where a rather large discrepancy was observed in the comparison as shown in Fig. 3. The accuracy could be improved by the more detailed measurements, i.e. varying the temperatures in smaller steps than in the present experiment.

As the reference spectrum for normalization, an experimental time spectrum of the α -radioactivity of ^8Li implanted in pure Al was used. It allows us to avoid the systematic errors caused by the beam on/off operations, since no measurable diffusion effects were observed in the case.

The diffusion coefficients, extracted in this way, are systematically smaller by about 30% as compared to those measured by PFG (pulsed filed gradient)-NMR (Nuclear Magnetic Resonance) spin-echo technique [7], in considering the Li contents of the present sample (48.5 at.%Li) [8]. Since the diffusion coefficients of Li in LiAl are strongly dependent on the Li contents as shown in [7], more detailed systematic investigation would be necessary in order to understand the discrepancy.

4. Conclusions and outlook

We have performed an experiment to measure the diffusion coefficients in LiAl compound by using a short-lived α -particle emitter, ^8Li , as a radiotracer. We have found that the time-dependent yields of the α -particles from diffusing ^8Li , primarily implanted in the sample, could be used as a measure of the diffusivity of the tracer in a non-destructive way. The time-dependent α -particle yields have been simulated for various diffusion coefficients, by taking account of the experimental conditions. Through the preliminary comparisons between the simulated and the experimental time spectra, the diffusion coefficients have been extracted with an uncertainty of 25%. The uncertainty is largely caused by the accuracy of the range of ^8Li in the sample, which is estimated by the SRIM-2003 code. On the other hand, the

present method could be applied as a range meter of Li isotopes in the compound where the Li diffusion coefficients are well known.

In our future facility, TRIAC, the quality of ^8Li beam will be improved, which gives narrower concentration-depth profile, at least by a factor of 4, as compared with the present experiment using the energy degrader. More than 100 times of ^8Li beam could be available, which allow us more detailed measurements in the limited time of experiment.

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