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DIFFUSION OF ^{6}Li IN TUNGSTEN AND TANTALUM[1]

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The objective of this work was the study of ^{6}Li diffusion in the Ta and W refractory metals. The samples were prepared by ion implantation of the 380 keV $^6Li^+$ ions into W and Ta thin foils up to the fluence of 10^{16} cm^{-2} and annealed up to the temperature 1900°C. The depth profiles of ^{6}Li were determined using the Thermal Neutron Depth Profiling (TNDP) technique. The results showed that diffusion of ^{6}Li in both W and Ta is very complex and cannot be described by simple Ficks laws. The Fickian diffusion is affected by the presence of traps and radiation defects in the sample surface layer. Further (and more detailed) TNDP study and computer simulations are therefore necessary.

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1 Introduction

The on-line isotope separation techniques (ISOL), for the production of ion beams of short-lived radionuclides [1], requires the fast separation of nuclear reaction products from irradiated targets, followed by their transfer into an ion source. As a first step in this transport chain, the fast and efficient release of nuclear reaction products from the target material is of crucial importance. This very complex process comprises diffusion of the reaction products to the target surface and their subsequent evaporation (effusion) from the target surface [2].

Reliable data about the diffusion of the elements of interest are crucial for the optimization of ISOL experiments from the point of view of the release time and efficiency. The release of the radioactive products from the target materials was extensively studied e.g. in [3]. The diffusion coefficients of several elements in carbon and different metals were determined by fitting of the release curves. Since this approach deals with a combination of two processes, i.e. diffusion and effusion, the data on the diffusion coefficients obtained in this way are model-dependent. Direct measurement of the diffusion (under the conditions met in the ISOL targets) is of interest since it will distinguish the role of product diffusion and effusion in the release process. Reliable data on Li isotope diffusion in ISOL target materials are scarce, see e.g. [4–6]. We started a project to study the diffusion of Li in potential ISOL target materials: Nb, Mo, Rh, Hf, Ta, W, Re, Ir, Pt, TaC, WC including various types of carbon. In this work results on diffusion of ^{6}Li in tungsten and tantalum are presented (see also [7]).

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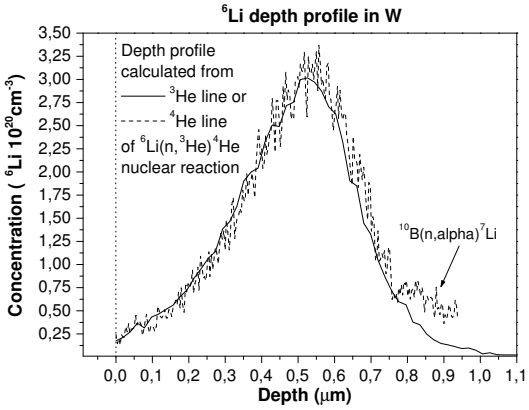

Fig. 1. Typical depth profile of ^{6}Li implanted with the energy 380 keV into W to the fluence 1×10^{16} cm^{-2}. The depicted profiles were calculated from both ^{3}H and ^{4}He lines of the ^{6}Li(n$_{th}$,^{3}H)^{4}He nuclear reaction. In the energy spectrum ^{10}B contamination was also observed.

2 Experimental

The samples were produced by the implantation of 380 keV ^{6}Li$^+$ ions into polycrystalline W and Ta foils of 50 μm thickness to the fluence 1×10^{16} cm^{-2}. Then the samples were annealed under the vacuum below 10^{-4} mbar at the temperatures from 1000 – 1900 °C. The ^{6}Li depth profiles on as implanted and annealed samples were determined by the Thermal Neutron Depth Profiling technique (TNDP) [8], making use of ^{6}Li(n$_{th}$,^{3}H)^{4}He nuclear reaction. The TNDP technique is based on the (n,p) or (n,α) exothermal nuclear reactions, induced by thermal neutrons, which exist on certain isotopes of several (light) elements (e.g. ^{3}He, ^{6}Li, ^{7}Be, ^{10}B, etc). TNDP provides an isotope specific, non-destructive technique for the measurement of concentrations versus depth distributions in the near-surface regions of the solids. It is an ideal analytical tool for diffusion studies for light elements in solid materials.

The samples were irradiated with thermal neutrons (with a 10^7 n$_{th}$ cm^{-2}s^{-1} neutron flux) from the 6 m long neutron guide installed at LWR-15 nuclear reactor (in NRI Řež). The charged reaction products were registered by semiconductor detectors (Hamamatsu, PIN diode) and the ^{6}Li depth profiles were calculated from the measured energy spectra using standard procedure and SRIM [9] stopping power data. In the present experimental arrangement the ^{6}Li detection limit is about 10^{13} at. cm^{-2} and the depth resolution varies from 15 nm (^{4}He) to 75 nm (^{3}H).

3 Result and discussion

The as implanted ^{6}Li depth profiles exhibit a typical Gaussian like shape with small left-hand side asymmetry for both W and Ta substrates, see an example in Fig. 1. In what follows the

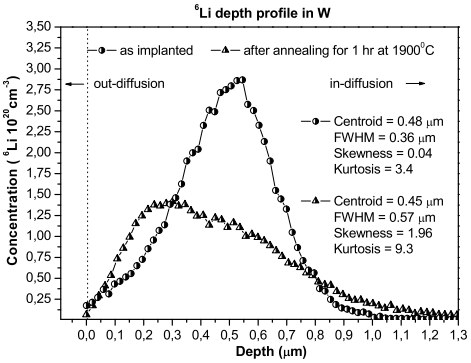

Fig. 2. Depth profiles of ^{6}Li (implanted into W with the energy 380 keV and fluence 1×10^{16} cm^{-2}) as implanted (circle) and after annealing for 1 hr at the temperature 1900°C (triangle up).

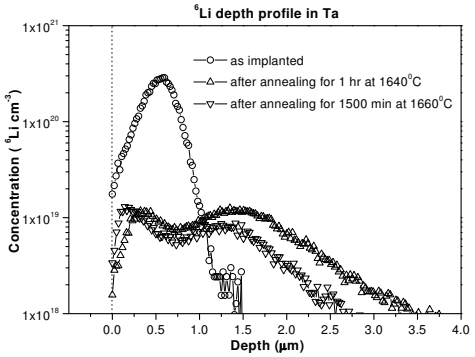

Fig. 3. Depth profiles of ^{6}Li (implanted into Ta with energy 380 keV and fluence 1×10^{16} cm^{-2}) as implanted (circle), after annealing for 1 hr at the temperature 1640°C (triangle up) or for 1500 min at the temperature 1660°C (triangle down).

profile position, width, asymmetry and curvature are characterized by projected range R_P, range straggling ΔR_P (1$^{\rm st}$ and 2$^{\rm nd}$ moments of the profile), skewness and kurtosis (normalized 3$^{\rm rd}$ and 4$^{\rm th}$ moments). The measured projected ranges (R_P ≈510 and ≈590 nm for W and Ta, respectively) and range stragglings (ΔR_P ≈200 nm and ≈240 nm for W and Ta, respectively) agree within experimental uncertainties with those calculated with the SRIM code [9].

The annealing is expected to result in the broadening of as implanted depth profiles due to diffusion of the implanted Li atoms from their initial positions to the sample bulk, and towards the sample surface as well. In the present case the diffusion could, however, be affected by the radiation defects generated by the high fluence implantation in the sample surface layer.

The results of hitherto performed measurements can be summarized as follows:

^{6}Li in W. The 1-hour annealing, at temperatures below 1560°C, does not lead to a measurable broadening of the Gaussian shaped ^{6}Li depth profile. For the annealing at temperatures above 1640°C, the significant broadening (Δx) of the depth profile is observed, from which the diffusion coefficient D can be estimated using common relation $2 \cdot D \cdot t = \Delta x^2$ (t being the annealing time). The diffusion coefficient varies from $D = 2 \times 10^{-14}$ to 1×10^{-13} cm^2 s^{-1} in the temperature interval from 1640 - 1900°C. The activation energy estimated from the Arrhenius plot is about 1.5 eV. The as implanted profile obeys Gaussian like shape with low skewness (0.04) and kurtosis close to 3. The annealing at 1900°C for 1 hour leads to both otward and inward diffusion of ^{6}Li (see Fig. 2) which cannot be described by a simple Fickian mechanism (as could be expected). The profile increases but the profile centroid remains practically unchanged.

^{6}Li in Ta. The 1-hour annealing at temperatures below 1560°C does not lead to a measurable broadening of the Gaussian shaped ^{6}Li depth profile in analogy with ^{6}Li behavior in W. At much longer annealing times ($>$ 25 hour) a slow diffusion of ^{6}Li atoms towards the sample surface is observed even at lower temperatures. In this case, the resulting profile acquires large left hand asymmetry with a significant part (about 10%) of the ^{6}Li atoms being transferred from the

implantation site toward the sample surface.

Annealing to temperatures above 1640°C for 1 hour of the samples implanted to the fluence of 1×10^{16} cm^{-2} results in a dramatic change of the profile shape. Fast inward diffusion of the ^{6}Li atoms is observed and the resulting profile exhibits two concentration maxima, one near the sample surface and another, less pronounced, at the depth of about 1.5 μm. The diffusion behavior is illustrated in Fig. 3. The shape of the diffusion profile in the region from 1.5 – 3.5 μm indicates clearly a capturing of ^{6}Li atoms on traps formed at defects produced by ion implantation. The ^{6}Li atoms may combine with the traps and create immobile complexes. The nature of the defects (vacancies, dislocation loops, etc.) can not be revealed only by the TNDP method.

4 Conclusion

The depth profiles of the ^{6}Li atoms implanted into W and Ta and annealed to different temperatures were studied by the TNDP technique for the first time. First results indicate a complex behavior of ^{6}Li atoms implanted into W and Ta, which can be affected by the presence of radiation defects or other traps in the sample surface layer. To obtain more definite information on the ^{6}Li diffusion in W and Ta, much richer experimental data are needed. The further experiments and computer simulations, taking into account the possible effects of trapping centers and spatially dependent diffusion coefficients, are therefore in progress.

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