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journal or publication title	Science reports of the Research Institutes, Tohoku University. Ser. A, Physics, chemistry and metallurgy
volume	35
number	2
page range	237-245
year	1991-03-05
URL	http://hdl.handle.net/10097/28342

Neutron Irradiation Effects in Al-Li and Al-Mg-Li Alloys

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(Received January 29, 1991)

Synopsis

Al-2.3wt%Li and Al-4.1wt%Mg-1.1wt%Li alloys were neutron-irradiated in JOYO, JMTR and RTNS-II with different types of energy spectra. The changes of He production by different neutron sources and of precipitates due to the isothermal annealing at 400°C were investigated by transmission electron microscopy (TEM). It was confirmed from TEM observations that He production by transmutation and the coagulation of He atoms after the annealing depend on neutron energy spectra used for the irradiation. It was also found that the addition of Mg to Al-Li alloy strongly suppresses the growth of He bubbles. Interrelations among precipitates, vacancies, He atoms and Li atoms were investigated, obtaining a reasonable coincidence between mutual amounts.

I. Introduction

Al-Mg-Li alloys have been developed as materials with low residual radioactivities after neutron irradiation [1-3]. Present authors have investigated on the changes of the mechanical properties after neutron irradiation [4,5]. For practical use of those materials, He bubbles anticipated to be formed from transmutation under neutron irradiation [6,7] give serious influences on mechanical properties such as void swelling, He embrittlement, etc. It is important for such research to consider not only the effects of the fluence, but also the energy spectrum of the neutrons used, since the type of radiation damage strongly depends on the neutron energy. However, the effects of the energy spectrum have not yet been studied fully.

Therefore, the present paper is conducted to study the effects of different

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energy spectra on He production and also the effect of the addition of Mg to Al-Li alloy on the formatiuon of He bubbles.

II. Experimental Procedures

Specimens used were Al-Li and Al-Mg-Li alloys. Their chemical compositions were Al-2.3wt%(8.3at.%Li) and Al-4.1wt%Mg-1.1wt%Li(-4.1at.%Li) alloys. The Al-Mg-Li alloy was the same as the specimens used in the previous works [4,5]. The fabrication method, the physical properties and microstructures after processing, and impurity contents of these alloys have been reported also in the previous papers [1,2,6]. The alloys were cold-rolled to 0.15mm thickness for Al-Mg-Li alloy and to 0.5mm thickness for Al-Li alloy. TEM disks of 3mm diameter were annealed at 500°C for 24 hours in Ar gas atmosphere before the neutron irradiation.

Specimens were irradiated in facilities with different neutron energy spectrum, namely, JMTR, JOYO and RTNS-II. Typical spectra of JMTR and JOYO are shown in figs.1 (A) and (B) [8,9]. Experimental conditions such as the fluence of fast neutrons (E_n>0.1MeV), total fluence (E_n>0.065eV) and irradiation temperatures are tabulated in table 1. In order to clarify the annealing behavior of irradiation-induced defects, and precipitates, a part of specimens were annealed at 400°C for 2, 5 and 10 hours in an electric furnace of Ar gas atmosphere after vacuum pumping.

TEM observations were carried out for as-irradiated specimens and after isothermal annealings at 400°C. All specimens were thinned by electropolishing in a 20% perchloric acid solution in ethyl alcohol at 0°C using a Tenupol apparatus at 20V and 50mA, and then examined by a JEOL 200-CX electron microscope operating at an accelerating voltage of 200kV.

III. Experimental Results and Discussion

1. Neutron energy spectrum

On study of the neutron irradiation defect, it should be noted that thermal neutrons play an important role in the He production by (n,α) reaction of target materials. Namely, the production of the He atoms from Li, Al and Mg elements are expected. As seen in Figs.1 (A) and (B), neutron spectra of JMTR and JOYO are different, the former having a neutron energy peak in the thermal region. From these spectra, we can simulate the amounts of He induced by the nuclear transmutation in Al-Li and Al-Mg-Li alloys irradiated in JMTR and JOYO. From each energy spectrum, average displacement cross section and He production cross section for ^{6}Li, ^{7}Li, ^{26}Mg and ^{27}Al were estimated [10].

Table 2 shows the total displacement damage (dpa) and the He production (appm) calculated with our experimental conditions. For the calculation for RTNS-II irradiation, we assumed a single spectrum with an energy of 14.0 MeV. although the neutron energy distributes in the region of 13.5-14.5MeV, Amount of He produced from Mg, Al and Li for the irradiation fluences were independently estimated. And their sum for each alloy is shown in table 2.

2. JMTR neutron irradiation

In order to study the effect of Mg addition on the formation of He bubbles, Al-Li and Al-Mg-Li alloys were irradiated in the JMTR reactor up to a total fluence of $4.6x10^{24}n/m^2$ at 250°C.

Fig. 2 shows typical TEM bright field images for as-grown (A,B) and after annealing at 400°C for 10 hours (C,D) of Al-Li and Al-Mg-Li alloys, respectively. In Al-Li alloy (Fig.2(A)), He bubbles of approximately 13nm in diameter with a density of $4x10^{19}/m^3$ were observed as small white spots. However, in Al-Mg-Li alloy (Fig.2(B)), no He bubble was observed. As seen in Fig. 2(C), He bubbles in the Al-Li alloy grew in size to an average diameter of 57nm after annealing at 400°C for 10 hours. It should be noted that they seemed to be trapped on dislocations. Fig. 2(D) shows He bubbles and precipitates in Al-Mg-Li alloy. It should be noted that He bubbles agglomerated surrounding precipitates. After the annealings for 2 or 5 hours, He bubbles were observed along dislocations. Irregular shaped images and spherical images in Fig. 2(D) are precipitates. Especially, the latter were identified as Al_3Li in the previous paper[5).

Annealing behavior of He bubbles induced by the neutron irradiation and the correlation between the precipitates and bubbles on annealing were investigated. Fig. 3 shows the relationship between the size of He bubbles and annealing time for Al-Li and Al-Mg-Li alloys. The curve for the size of precipitates vs. the annealing time in Al-Mg-Li alloy is also inserted in this figure. In this figure, we can see that the size of He bubbles in Al-Li alloy is larger than that of Al-Mg-Li alloy, and that the bubble sizes of both alloys increase with increasing annealing temperature. The size of precipitates against annealing temperature in the Al-Mg-Li alloy showed a similar tendency.

Fig. 4 shows the relation between the swelling, which is defined as product of density and volume of bubble, and annealing time. As shown in the table 2, the estimated amount of He atoms induced by nuclear transmutation were 892appm for Al-Li alloy, and 439appm for Al-Mg-Li alloy, after the consideration of the amounts of Li in both alloys. Therefore, the ratio of the He production rates of the Al-Li and the Al-Mg-Li should be 2. However, as seen in fig. 4 the ratio of swelling was approximately 8 experimentally after the 10 hours annealing. From this fact, it is obvious that the addition of Mg atoms to Al-Li alloy suppresses strongly the swelling. This is the advantage of the Al-Mg-Li alloys over the Al-Li alloys to be used as a reactor material.

3. JOYO neutron irradiation

In JOYO, Al-Mg-Li alloys were irradiated at 400±10°C. Fig. 5 shows typical microstructure after irradiation fluences of $1.5x10^{24}n/m^2$ (A) and $1.3x10^{26}n/m^2$ (B).

Dislocation loops and AL_3Li precipitates identified in the previous paper[5] are also seen in fig. 5(A). The loops grew with increasing fluence and eventually vanished. At the highest fluence of $1.3x10^{26}n/m^2$, voids appeared as seen in fig. 6(B). Their density and size were about $2.4x10^{19}/m^3$ and 37nm, respectively. In a specimen irradiated to a fluence of $1.5x10^{24}n/m^2$, they were not observed after annealing at 400°C for 5 hours.

Fig. 6 shows the relationships between the density of Al_3Li and neutron fluence, and also between the size of precipitate and fluence. It should be noted that the density of precipitate decreased with an increase of the fluence, and the size of precipitate increased in contrast to the former.

The correlation between the void and the Al_3Li precipitate is discussed below. Table 3 shows the densities of the voids and Al_3Li precipitates, and their sizes after irradiation up to a fluence of $1.3x10^{26}n/m^2$. Assuming that the volume of voids are occupied by cluster of vacancies which has the volume of Al atom, the number of vacancies in unit volume (Vn) can be calculated from the density and the size of void measured experimentally, namely,

$$V_n = \rho_1(4\pi/3)\cdot(D_1/2)^3N_{Al} \quad . \qquad (1)$$

Where ρ_1 , D_1 and N_{Al} are the density of void, diameter and the number of Al in unit volume, respectively. We adopted the values shown in table 3 for ρ_1 and D_1, and a value of $6x10^{28}/m^3$ for N_{Al} . The value of $3.8x10^{25}/m^3$ for V_n was obtained from the eqn. (1).

From the changes of density and size of Al_3Li precipitates, we can also calculate the number of Li atoms (ΔN_{Li}), which have moved to form precipitates as

$$\Delta N_{Li} = \Delta\{\rho_2(4\pi/3)\cdot(D_2/2)^3\}N_{Li} \quad , \qquad (2)$$

where ρ_2 , D_2 and N_{Li} are density of precipitates, diameters of precipitates, and the number of Li atoms per unit volume in Al_3Li precipitates, respectively. We adopted the values shown in table 3 for ρ_2 and D_2 , and $1.6x10^{28}/m^3$ for N_{Li} . The value of $2.5x10^{25}$atoms/m^3 for ΔN_{Li} was obtained from eqn. (2).

The values obtained from eqns. (1) and (2) showed a good correlation to each other. From this result, it can be concluded that the formation of void may be due to the coalescence of vacancies which detrapped from impurity atoms. The growth of precipitates of Al_3Li may be promoted by Li atoms freed from vacancies.

4. RTNS-II neutron irradiation

TEM observations of Al-Mg-Li alloys irradiated at 200°C up to a fluence $2.0x10^{22}n/m^2$ and after annealing at 400°C for 5 hours were carried out. However, no cavity was observed even though the irradiation had been carried out by neutrons with very high energy. As is obvious from table 2, in RTNS-II irradiation the values of displacement per atom (dpa) and He production (appm) are very low in comparison with those of JMTR and JOYO. Namely, the values of 0.01 dpa and 0.48

appm in RTNS-II may be too low to observe the cavity by TEM.

IV. Conclusions

Al-2.3wt%Li and Al-4.1%Mg-1.1%Li alloys were irradiated in JOYO, JMTR and RTNS-II with different types of energy spectra. The changes of He production and of precipitates due to the isothermal annealing at 400°C were investigated by TEM. The main conclusions obtained with the TEM observations are summarized as follows.

1) The difference of He production depending on the neutron energy spectrum were confirmed experimentally.

2) The addition of Mg to Al-Li alloys strongly suppressed the formation of He bubbles.

3) For Al-Mg-Li alloys after JOYO neutron irradiation, the relationship between voids and Al_3Li precipitates were examined. The vacancy concentration showed a good correlation with the number of Li atom contributed to the formation of Al_3Li precipitates.

Acknowledgements

The authors wish to express their cordial thanks to Oarai Branch of Tohoku University for facilitating post-irradiation experiments after JOYO, JMTR and RTNS-II neutron irradiations. We are also grateful to Dr. T.Yoshiie for help with the irradiation in LLNL and the arrangements for the post irradiation procedure. Al-Mg-Li alloys were supplied by Sumitomo Light Metal Industry.

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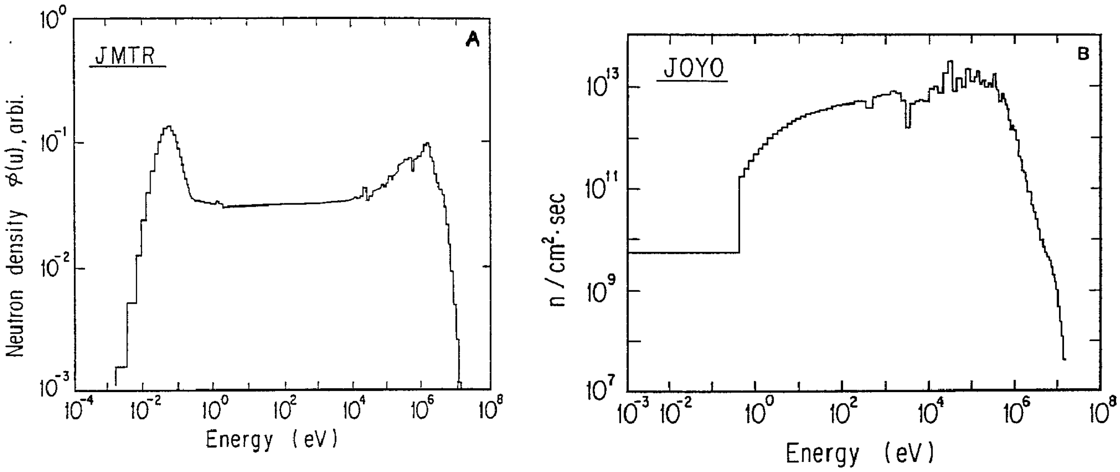

Fig. 1. Typical neutron energy spectrums of JMTR and JOYO used in the irradiation. A; JMTR B; JOYO.

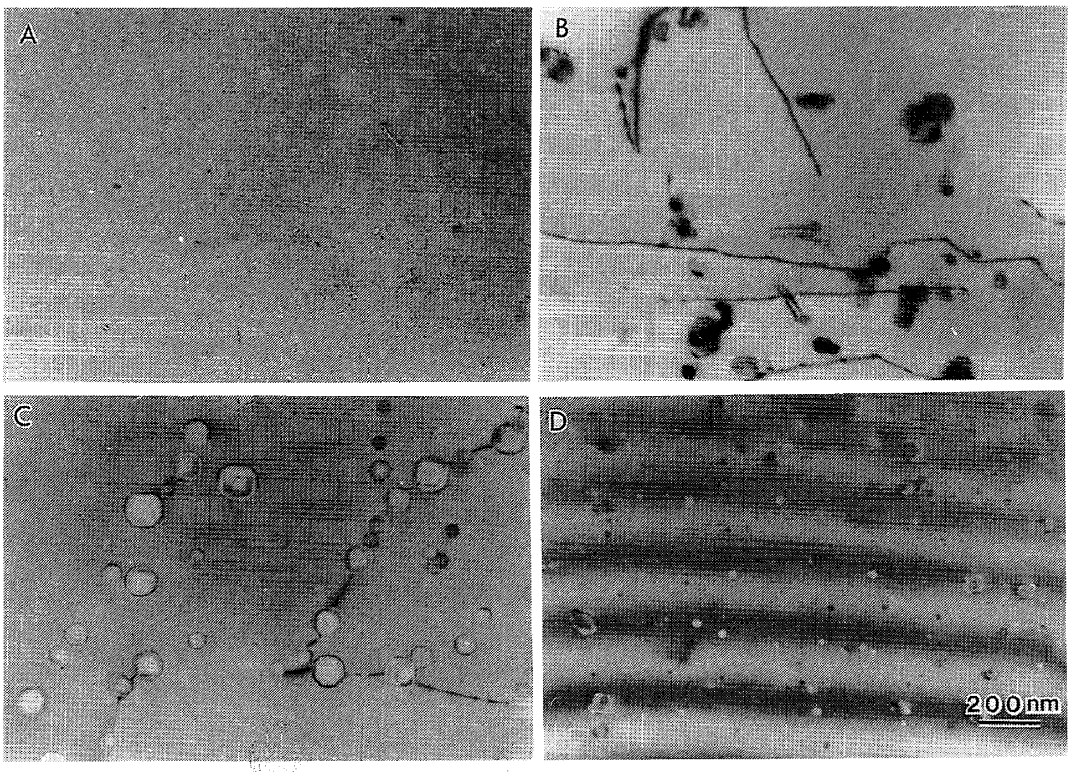

Fig. 2. Comparison of microstructure before (A,B) and after annealing at 400°C for 10 hours (C,D) of Al-Li and Al-Mg-Li alloys irradiated in JMTR at 250°C. The total fluence was $4.6 \times 10^{24} n/m^2$. A and C; Al-Li alloy, B and D; Al-Mg-Li alloy.

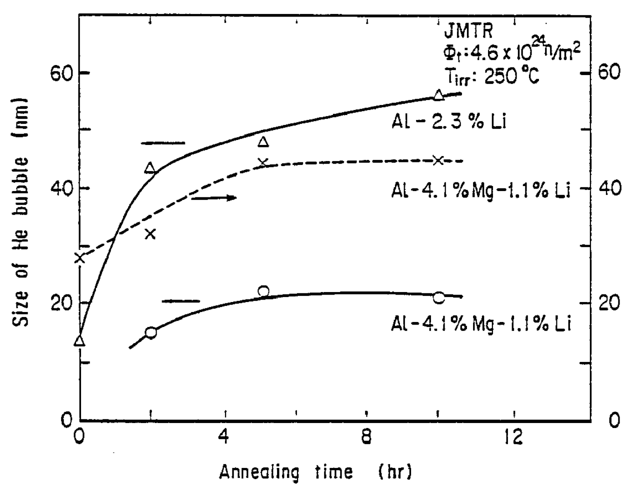

Fig. 3. Variation of sizes of He bubble and precipitate as a function of annealing time at 400°C. (JMTR; $4.6x10^{24}n/m^2$ at 250°C).

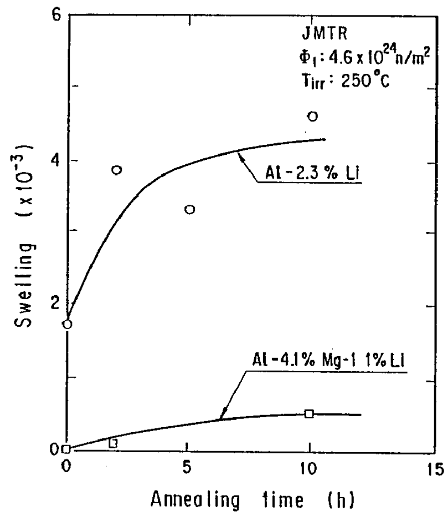

Fig. 4. Comparison of swelling in Al-Li and Al-Mg-Li alloys as a function of annealing time at 400°C. (JMTR;$4.6x10^{24}n/m^2$ at 250°C).

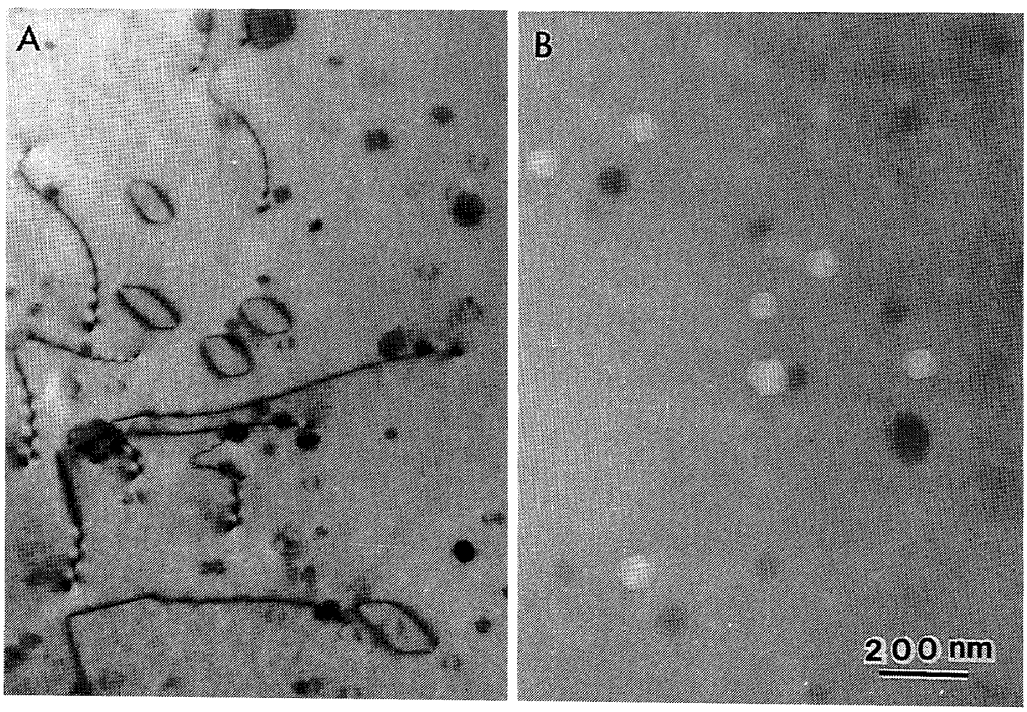

Fig. 5. Comparison of damage structure of Al-Mg-Li alloy irradiated in JOYO at 400°C. (A; $1.5x10^{24}n/m^2$, B; $1.3x10^{26}n/m^2$).

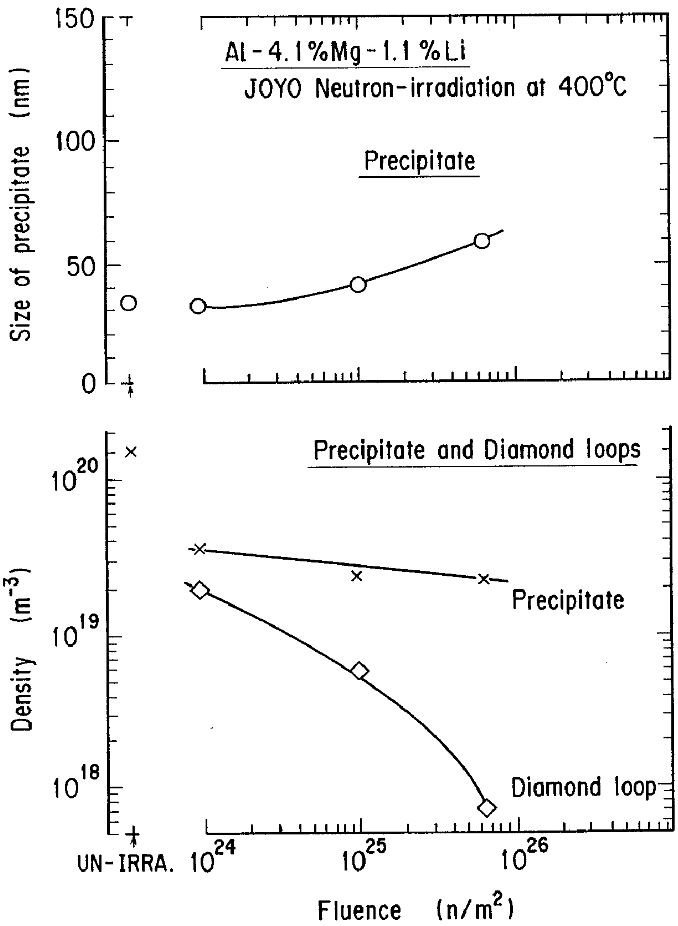

FIg. 6. Neutron fluence dependences of density and size of Al_3Li precipitates in Al-Mg-Li alloy irradiated in JOYO at 400°C.

Table 1. Irradiation conditions of Al-Li and Al-Mg-Li alloys used in the present work.

ϕ (n/m^2)	JMTR	JOYO			RTNS-II (14.1 MeV)
		# 1	# 2	# 3	
Total Fluence (E_n > 0.065 eV)	4.6×10^{24}	1.5×10^{24}	1.5×10^{25}	1.3×10^{26}	2×10^{22}
Fluence (E_n > 0.1 MeV)	2.4×10^{24}	7.6×10^{23}	7.5×10^{24}	6.3×10^{25}	
Irrad. Temp. (°C)	250	400			200

Table 2. The estimated values of displacement per atom (dpa) and He production (He appm) for Al-Li and Al-Mg-Li alloys.

Facilities	Al-Li Alloy		Al-Mg-Li Alloy	
	Displacement per atom (dpa)	He production (appm)	Displacement per atom (dpa)	He production (appm)
JMTR	0.25	892	0.25	439
JOYO #1	0.06	5.4	0.06	2.7
JOYO #2	0.62	53	0.62	26
JOYO #3	5.17	430	5.23	212
RTNS-II	0.01	0.48	0.01	0.48

Table 3. The changes in the density and the size of voids and Al_3Li precipitates in Al-Mg-Li alloy irradiated in JOYO at 400°C.

Neutron fluence	$1.5 \times 10^{24} n/m^2$	$1.3 \times 10^{26} n/m^2$
Density of void (ρ_1)	———	$2.4 \times 10^{19} m^{-3}$
Size of void (D_1)	———	37 nm
Density of Al_3Li (ρ_2)	$3.5 \times 10^{19} m^{-3}$	$2.2 \times 10^{19} m^{-3}$
Size of Al_3Li (D_2)	32 nm	57 nm