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Void formation in an Al-Mg-Si alloy under different precipitation conditions after irradiation with 100 keV Al ions

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The influence of different precipitation treatments upon the radiation-induced void formation was studied on an Al–Mg–Si alloy and the results were compared with those obtained on high-purity Al. The changes in the microstructure and the void formation were investigated by TEM methods for the dose range 0.8 to 80 dpa and the temperature range 55 to 250° C. The irradiations were carried out using 100 keV Al ions. High-purity Al showed void formation over the whole temperature range investigated with a maximum volume increase $\Delta V/V$ of 3.4% at 150° C. The behaviour of the Al–Mg–Si alloys depends strongly upon the thermal pretreatments. The alloy in its homogenized state shows no voids between 55 and 140° C. Trapping of vacancies on solute Si atoms could be the explanation. An ageing treatment leading to coherent precipitates results in the complete suppression of void formation for the temperature region of highest swelling ($0.35\ T_s \leqslant T \leqslant 0.44\ T_s$). On the other hand, treatments which cause incoherent or partially coherent precipitates result in swelling. The amount of swelling, however, is lower compared to pure Al and the temperature of the swell maximum is shifted to lower temperatures. These results could be explained by the mechanism of trapping vacancies by solute atoms, on the one hand, and the coherent precipitates acting as recombination centres, on the other.

1. Introduction

Previous work has shown that voids form and grow in aluminium and aluminium alloys in the temperature range 25 to 150° C after neutron fluences corresponding to ~ 0.1 displacements per atom (dpa) [1, 2]. Void formation and swelling has been found to be considerably reduced in neutron-irradiated aluminium alloys containing Mg_2Si particles or a certain amount of solute magnesium and silicon, respectively. However, very little information is known about the influence precipitates in an AlMgSi alloy have on the development of irradiation-induced microstructures at high damage levels.

In this paper experimental results from transmission electron microscopy (TEM) studies are presented. They were carried out to assess the irradiation damage characteristics of an AlMgSi alloy with different precipitation conditions after ion irradiations to displacement levels up to 80 dpa using 100 keV aluminium ions. Pure aluminium was also investigated in order to compare void swelling.

2. Experimental procedure

The pure aluminium and the AlMgSi alloy used were taken from rolled sheet of 0.2 mm thickness. Chemical analysis of the major elements (wt %) of

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TABLE I Chemical composition of pure aluminium (ppm) and aluminium alloy (wt%)

Material	Pure aluminium	AlMgSi
Si	< 2 ppm	< 0.61
Mn	< 1 ppm	< 0.005
Cr	< 1 ppm	< 0.001
Co	< 1 ppm	< 0.001
V	< 1 ppm	< 0.005
Fe	< 5 ppm	< 0.01
Cu	< 5 ppm	< 0.001
B		< 1–2 ppm
Mg	< 5 ppm	< 0.9
Zn	< 2 ppm	< 0.005

the materials is given in Table I. The AlMgSi samples were solution treated at 560° C for 4 h. Ageing treatments to obtain coherent and incoherent precipitates, respectively, were carried out either in vacuum or in oil baths.

The precipitation kinetics at different ageing temperatures were followed by resistivity measurements performed at $-195°$ C. This procedure guaranteed that the ageing times chosen led to the equilibrium state of the precipitates. Experimental details, i.e. specimen shape, quenching conditions, heat treatment and methods of measurement have been described previously in more detail [3].

The irradiations with 100 keV Al^+ ions were performed using the GSI (Gesellschaft für Schwerionenforschung, Darmstadt) Duopigatron Ion-Source [4]. Temperatures between 55 and 300° C were chosen, and the accumulated displacement damage ranged from 0.08 to 8×10^{16} ions cm^{-2} which corresponds to 0.8 to 80 displacements/atom.

Representative electron micrographs were taken of each specimen examined, and evaluation of the average void diameter, void concentration volume fraction and dislocation density was made using methods described elsewhere [3].

3. Results

3.1. The temperature dependence of swelling in pure aluminium

Pure aluminium was irradiated at 55, 100, 150, 190, 250 and 300° C. The average void diameter, void concentration, volume fraction and dislocation densities are shown in Table II. The void swelling is plotted in Fig. 1. Representative micrographs of void structures are shown in Fig. 2. It can be seen that void swelling increases from that attained at 55° C to reach a maximum at about 150° C. Void concentration at 250° C was found to be negligible. No voids could be observed at all at 300° C.

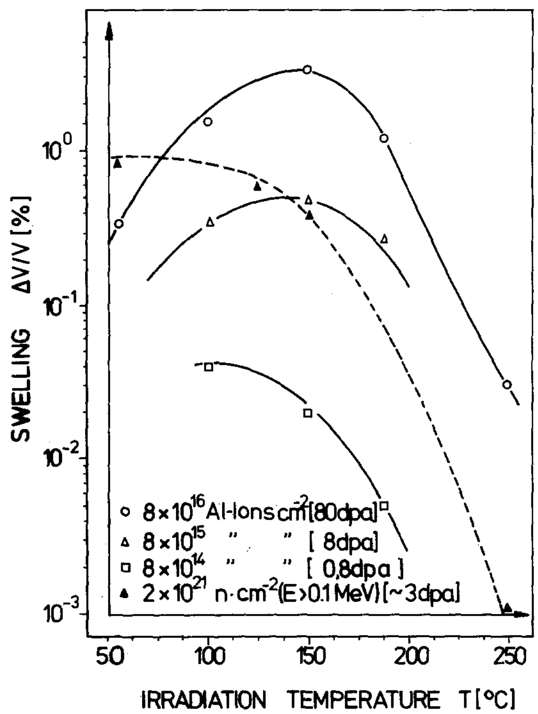


Figure 1 Void swelling as a function of temperature in pure aluminium after irradiation with 100 keV Al^+ ions. Swelling after neutron irradiation is shown for comparison.

TABLE II Results of 100 keV Al^+ ion irradiation in pure aluminium. Ion dose 8×10^{16} cm^{-2}

Irradiation temperature (° C)	Mean void diameter (Å)	Swelling $\Delta V/V$ (%)	Void concentration (10^{15} cm^{-3})	Dislocation density (10^{10} cm^{-2})
55	105	0.35	4.9	4
100	168	1.5	4.7	3
150	525	3.37	0.39	2.8
190	1040	1.22	0.02	1
250	783	0.03	0.001	0.3
300	–	–	–	–

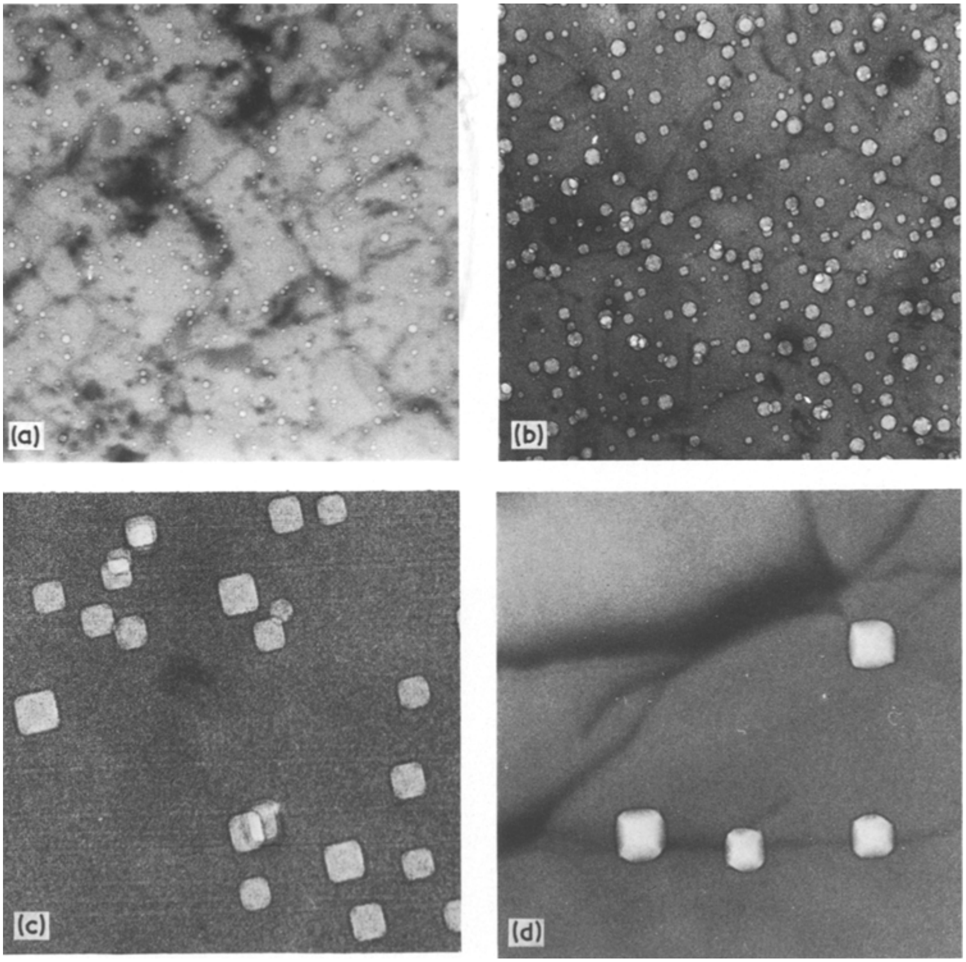


Figure 2 Voids in pure aluminium after irradiation with 100 keV Al^+ ions at a dose of 8×10^{16} cm^{-2} as a function of temperature. (a) 55° C, (b) 100° C, (c) 150° C, (d) 190° C.

3.2. The influence of different precipitation treatments upon void formation in an AlMgSi alloy

The void swelling behaviour is shown to be strongly dependent upon thermal pretreatment. The alloy in the homogenized state shows no voidage between 55 and 140° C. However for the temperature region of 0.35 to 0.45 T_s, ageing treatment leading to coherent precipitates results in a complete suppression of void formation. Pretreatments which cause incoherent or partially coherent precipitates, however, cause swelling but only after irradiation at a total displacement damage of 80 dpa. No voids at all could be observed after irradiations for a total displacement damage of 0.8 dpa. The results of the investigations carried out are given in Table III. Examples of void formation and dislocation structure of AlMgSi are shown in Figs. 3 and 4. The void swelling is plotted in Fig. 5 as a function of irradiation temperature.

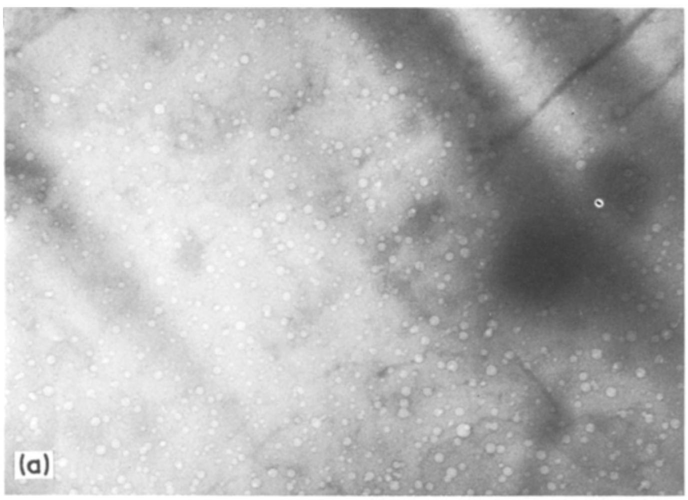
4. Discussion

4.1. Void formation in aluminium

The results show that voids can be produced in pure aluminium by 100 keV Al^+ ions within the temperature range 50 to 250° C. Void swelling is found to reach a maximum of 3.4% at ~ 150° C at a total displacement damage of 80 dpa. The temperature at which the maximum swelling occurs under Al^+ ion irradiation is about 70° C higher than that observed for aluminium irradiated with neutrons. This temperature "shift" is predicted by theory and is due to the fact that with Al irradiation the defect production rate is higher by about a factor of 1000 than during neutron irradation.

TABLE III Results of 100 keV Al^+ ion irradiation in AlMgSi at a temperature of 100 and 150° C. Ion dose 8×10^{16} cm^{-2}

Ageing condition	Irradiation temperature (° C)	Mean void diameter (Å)	Swelling $\Delta V/V$ (%)	Void concentration (10^{15} cm^{-3})	Dislocation density (10^{10} cm^{-2})
Annealed	100	–	–	–	3.5
and quenched	150	–	–	–	2.5
Aged at	100	–	–	–	3
140° C	150	–	–	–	2
Aged at	100	120	0.7	6.6	3
200° C	150	247	0.21	0.2	2
Aged at	100	103	0.3	4.0	3
260° C	150	232	0.08	0.1	1



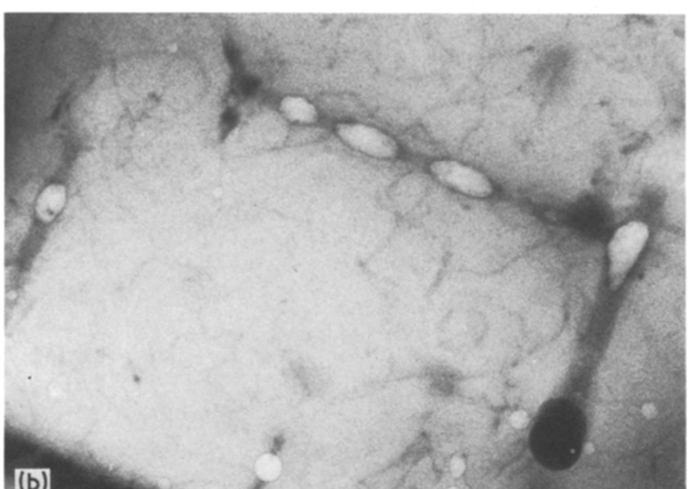


Figure 3 Typical void distributions observed in AlMgSi aged at 260° C irradiated to 8×10^{16} Al^+ ions cm^{-2} at the following temperatures: (a) 100° C; (b) 140° C.

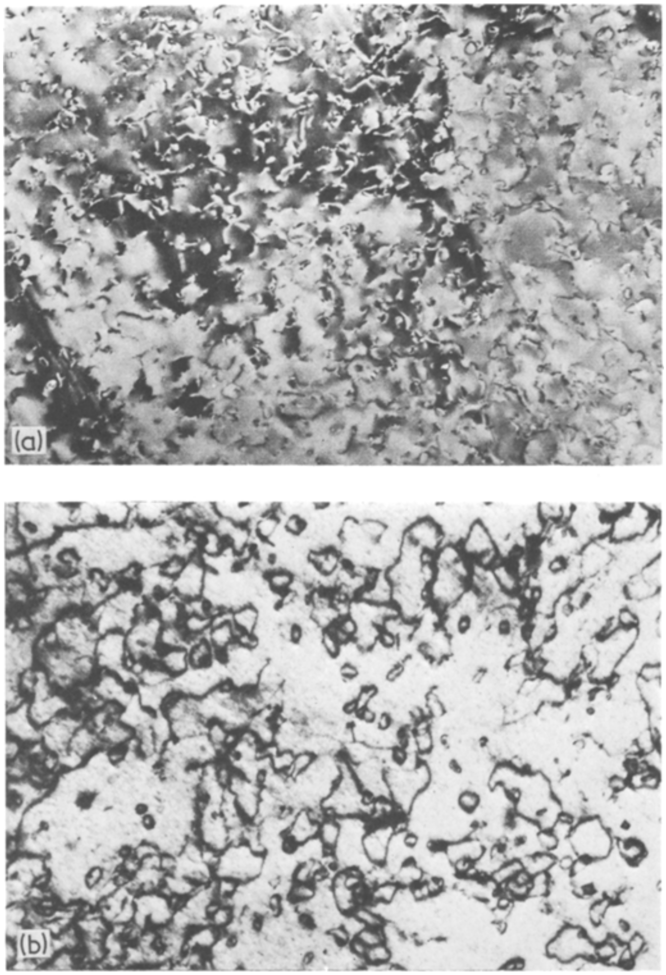


Figure 4 Typical dislocation structure observed in AlMgSi irradiated to 8×10^{16} Al^+ ions cm^{-2} at 140° C (a) in the aged and quenched condition, (b) after ageing at 140° C with coherent precipitates.

4.2. Void formation in AlMgSi

The results of Al^+ ion irradiation of AlMgSi after different precipitation treatments at temperatures between 50 and 200° C up to 8×10^{16} ions cm^{-2} indicate a strong dependence of void swelling upon precipitate condition and solute solicon and/or magnesium concentration, respectively. Neutron-irradiated specimens of nearly the same composition have been investigated by Farrell and co-workers at the Oak-Ridge Laboratories [5–7] and by Sturcken *et al.* at the Savannah River Laboratory [8]. Farrell showed on fully hardened specimens that void formation could not be observed at all up to a fast fluence of 1.6×10^{22} n cm^{-2} ($E > 0.1$ MeV) at 55° C. In overaged specimens a low void formation occurred above 4×10^{22} n cm^{-2} ($E > 0.1$ MeV). Sturcken *et al.* investigated AlMgSi specimens from structural components. Voids were not observed up to a maximum fast fluence of 5.8×10^{21} n cm^{-2} ($E > 0.8$ MeV). However, previous reactor radiation experiments had the well known disadvantages that the precipitation structure on the material was insufficiently known before irradiation and that silicon and hydrogen are produced. With Al^+ ion irradiation, neither alloy composition nor nuclei formation were influenced through additional gas production; in addition, the precipitates of the investigated alloy (AlMgSi) were clearly identified before irradiation using transmission electron microscopy and resistometric methods.

The results of our investigation show that

(1) finely dispersed coherent precipitates *completely* prevent void formation in the AlMgSi alloy up to 8×10^{16} Al^+ ions cm^{-2};

(2) voids could not be observed in the solution heat-treatment, homogenized condition, i.e. when magnesium and silicon are completely in solution;

(3) void formation occurred at ageing conditions which had resulted in larger (5000 Å) rod-shaped

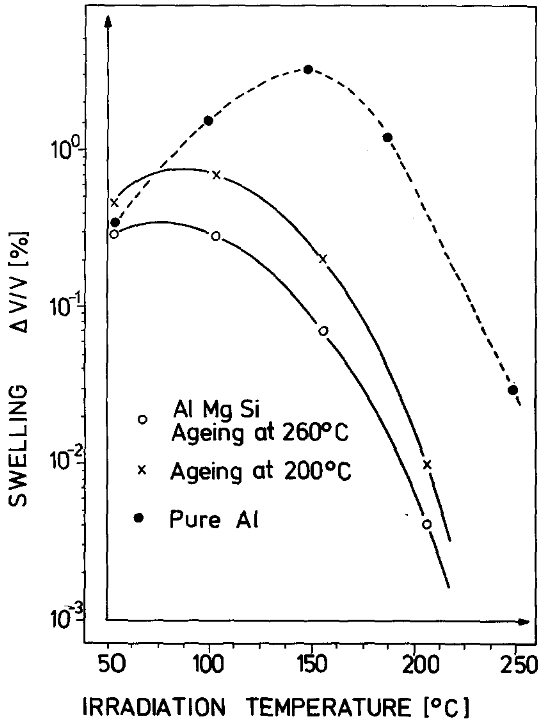


Figure 5 Void swelling as a function of irradiation temperature in AlMgSi with different precipitation conditions. Void swelling in pure aluminium is shown for comparison. (Displacement damage 8×10^{16} ions cm^{-2}).

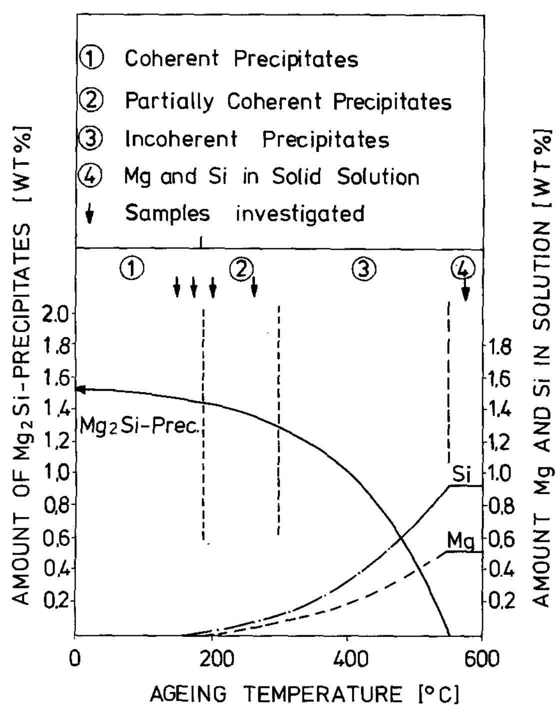


Figure 6 Solubility of silicon and magnesium and the degree of precipitation as a function of temperature. The irradiated precipitation conditions are indicated by arrows.

and partly incoherent precipitates. In addition, isolated large voids were found within large Mg_2Si precipitates.

When comparing the results of pure aluminium with the alloy AlMgSi in the solution-treated condition, one has to explain the total suppression of void formation in the latter alloy. The amount of Mg and Si in solid solution is 1.54 wt % (Fig. 6). According to a proposal of Smidt and Sprague [9] alloying elements in solid solution can trap vacancies and/or interstitials and thus appreciably increase the probability for recombination. A quantitative evaluation shows that by this mechanism void nucleation as well as void growth can be reduced. A trapping mechanism of this nature is conceivable in the case of the additive silicon as the binding energy of 0.25 eV [10] is sufficient to allow formation of a vacancy–silicon pair. Experimental results of Mayer and Morris [11] on AlSi alloys with varying silicon additions show that void formation can be suppressed with increasing silicon content. Such a mechanism can be assumed here with the AlMgSi alloy provided that silicon remains in solution during irradiation. TEM investigations have shown that precipitates are not formed. However, in the case of neutron irradiation where the time of exposure is about three orders of magnitude higher, i.e. at a certain damage level, it is questionable whether the formation of precipitates can be avoided.

In the case of magnesium additions it is difficult to argue in the same manner. The measured binding energies of < 0.1 eV [12] for a vacancy–magnesium complex are too small for a reduction in swelling. However, Mazey *et al.* [13] showed experimentally in simulation irradiations that the addition of magnesium to aluminium can reduce and suppress void formation as well. In this case it is possible to refer to the proposal of Venker *et al.* [14] who showed that the addition of fast diffusing elements might reduce swelling in binary nickel and copper alloys. Fujikawa and Hirano [15] measured the partial diffusion coefficient of Mg in Al and found that this value is higher than the corresponding self-diffusion coefficient of aluminium. The same argument might also hold for Si [16]. However, we have to point out that the application of this idea to this alloy requires further quantitative calculations.

In conclusion, the suppression of void formation in the AlMgSi alloy can be attributed to the elements magnesium and/or silicon in solution.

According to this model a pre-ageing treatment should reduce the ability to suppress void formation. From Fig. 6 one can see that the amount of silicon in solution after pre-ageing treatments at 140 and 170° C is lower than 0.01% compared with 0.03% after ageing at 200° C and 0.06% after ageing at 260° C. These thermal pretreatments lead to a maximum degree of precipitation. Consequently, a vigorous void formation should be observed: our results indicate a more complicated picture. Pre-ageing at 140 and 170° C, where fine dispersed coherent Mg_2Si precipitates are formed leads to a complete absence of void formation. In the case of ageing at 200 and 260° C, where according to our TEM investigations partially incoherent precipitates are formed, some swelling is observed. The absolute values are a factor of 3 to 4 lower than for the pure aluminium under identical irradiation conditions.

According to Brailsford and Bullough [17], reduced swelling in the presence of a higher density of coherent precipitates can be considered as the result of a trapping mechanism of vacancies at the coherent precipitate interfaces. Vacancies which move in this interface due to strain fields around these precipitates will be trapped and can recombine with the faster diffusing interstitial atoms. Thomas [18] found that the atoms in the Mg_2Si precipitates take up the same positions as in the matrix. According to his investigations, one layer zone including two magnesium rows and a silicon row has a width of 8.743 Å with the closest packing, and replaces in this manner three aluminium rows with a width of 8.586 Å. As a result of this atomic arrangement an expansion of about 2% vertical to the rod axis of the Mg_2Si precipitates can be found. This field of cylindrical compression can be observed by TEM. It can act as a trapping area for vacancies. For an explanation of the influence of the pre-ageing conditions it is necessary to obtain a quantitative figure about the integrated or total area of coherent interfaces for different heat treatments. A calculation of this parameter from the TEM data (concentration, size and type of interface) gives a value of 1×10^5 cm^2 cm^{-3} for pre-ageing at 140 and 1×10^4 cm^2 cm^{-3} for 200 and 260° C, respectively. This difference between the coherent interfaces (which can act as active trapping areas) is expected to be the reason for the different swelling behaviour.

5. Conclusions

The swelling behaviour of an AlMgSi alloy after irradiation with Al^+ ions can be explained by two important mechanisms:

(a) in the case of the solution-treated material the silicon in solid solution acts as a trapped centre for silicon–vacancy complexes and hence reduces swelling;

(b) in pre-aged conditions where nearly the whole amount of silicon and magnesium is bound in Mg_2Si precipitates the reduction of swelling is based upon the integral coherent interface between precipitates and matrix. This parameter is a function of the degree of dispersion of the second phase and its coherency. Vacancy trapping at these interfaces seems to be the important mechanism for reducing swelling.

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