Understanding of radiation effect on sinks in aluminum materials for research reactors

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1. Introduction

Aluminum and its alloy are widely used in structural materials for research reactor such as guide tube and cladding because of its physical properties such as high thermal conductivity, neutron economy and corrosion resistant properties [1]. Although aluminum and its alloy have excellent characteristic, radiation induced hardening and swelling are still important safety concern. In order to verify irradiation effect on aluminum and its alloy, irradiation behavior was examined at 328 K to fast (> 0.1 MeV) fluences up to 1.8 x 10^{27} n/m^2 which corresponding maximum dpa was 260 [2]. From this experimental research, radiation induced swelling and hardening was analyzed by various alloy type. It was confirmed that 6061-T6 (1 Mg, 0.6 Si, 0.28 Cu, 0.2 Cr) and 5052-O (2.5 Mg, 0.25 Cr) have good swelling resistance. Swelling of both alloys was less than 1 % however, 5052 Al show higher swelling resistance. In the case of radiation hardening, 0.2 flow stress of 6061 was increased by 45 % while that of 5052 increased by 400 %.

From microstructural analysis, it was confirmed that dislocation loop, void and precipitate are major sinks which induced swelling and hardening. Among these defects, precipitation such as Mg_2Si and Si were generated by reaction between alloy elements and transmutations. Therefore, radiation induced swelling and hardening can be predicted by analyzing these defect. However, quantitative analysis of these defects has not been done by computational tools. Therefore, it is unclear that specific mechanism of alloy element effects on the irradiation swelling and hardening in aluminum alloys.

Historically, radiation induced phenomena such as swelling, growth and hardening is simulated by Mean Field Radiation Damage Theory (MFRDT). From the MFRDT, reactions of irradiation defect and sink are calculated and then sink density is evolved at each type of sinks. From this research, irradiation effect on materials can be calculated quantitatively. In the early 1980s, many theoretical research, which is based on the MFRDT, had been done about irradiation swelling and hardening [3-5]. However, there is not general modeling which could explain about radiation effect on aluminum and its alloy. Therefore, the aim of this research is prediction of radiation induced swelling and hardening of aluminum and its alloy in research reaction condition by using computation research tools.

2. Approach

Radiation induced phenomenon is occurred in wide time range which is expended from pico-second to day. Therefore, various simulation tool should be applied for accurate prediction.

2-1 Multi-scale Modeling

In order to calculate defect generation rate, molecular dynamics (MD) can be used in pico ~ micro second time scale and then behavior of defect agglomeration and dissolution can be simulated by kinetic Monte Carlo (kMC) or MFRDT. From the parameters of defect such as size and number density, swelling and hardening rate can be obtained. Lastly, practical composition of structural materials can be simulated for safety analysis.

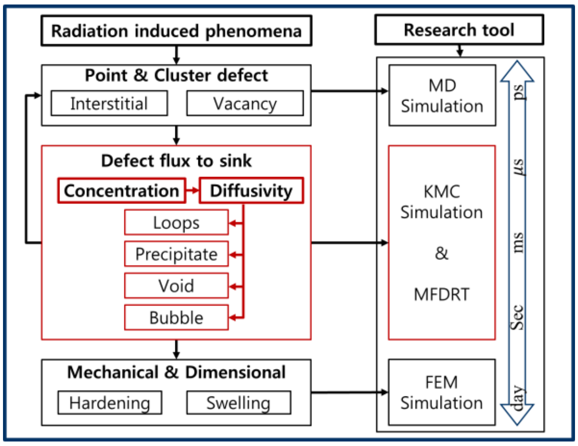

Fig. 1. Schematic of multi-scale modeling in radiation induced hardening

2-2 Methodology of MFRDT

Multi-scale modeling is very hard to simulate at once because this research spends a lot of time and fund. Therefore, in this research, firstly, MFRDT was conducted which could explain behavior of sink development and irradiation hardening and swelling. In case of MFRDT in aluminum, a slightly different approach is needed for the evaluation of radiation effect on aluminum and its alloy because neutron reaction with aluminum generate not only matrix defects but also transmutation (This behavior is unique phenomenon compared with other structure materials such as zirconium alloy and iron alloy).

Therefore, in order to simulate sink development including precipitation, simplified model is established with specific assumptions. In this research, it was assumed that

1. Reaction between defect and other atom happened in aluminum 1cm^3 matrix
2. Dislocation loop & void & precipitate are considered as major sink in aluminum matrix
3. Precipitate and void assumed that its morphology is sphere

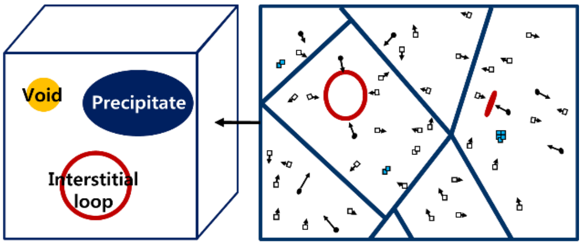

Fig. 2. Schematic of multi-scale modeling in radiation induced hardening

From this simplified assumption in MFRDT, defect concentrations are calculated by using defect rate equation. Owing to defect concentration and diffusivity, probability of defect reaction with sink could be calculated. After that, density change of sink can be analyzed by using initial sink density and reaction probability of sink. Therefore, it is very importation to model defect rate equation. Defect rate equations are expressed by Eqs. (1) and (2) defect generation rate and recombination rate are obtained from literature study [6].

$$\frac{dC_v}{dt} = K_o - K_{iv}C_iC_v - Z_v\rho C_v D_v \quad (1)$$

$$\frac{dC_i}{dt} = K_o - K_{iv}C_iC_v - Z_i\rho C_i D_i \quad (2)$$

K_0; Defect production rate
K_{iv}; Vacancy–interstitial recombination
ρ; Total sink density of aluminum matrix
Z_i and Z_v; Interstitial and Vacancy capture efficiency
C_i and C_v; Interstitial and Vacancy concentration
D_i and D_v; Interstitial and Vacancy diffusivity

Where, ρ is composed of density of major sink such as dislocation loops, precipitation, void and bubble in aluminum ($\rho = \rho_{DL} + \rho_{PPT} + \rho_{Void} + \rho_{Bubble}$). In this equation, density of sinks analyzed by number density and radius because radius could be calculated by probability of defect reaction with sink. However, in case of number density, there is no theoretical modeling to predict for irradiation. Therefore, experimental result is used for analysis of density change of sink,

3. Result

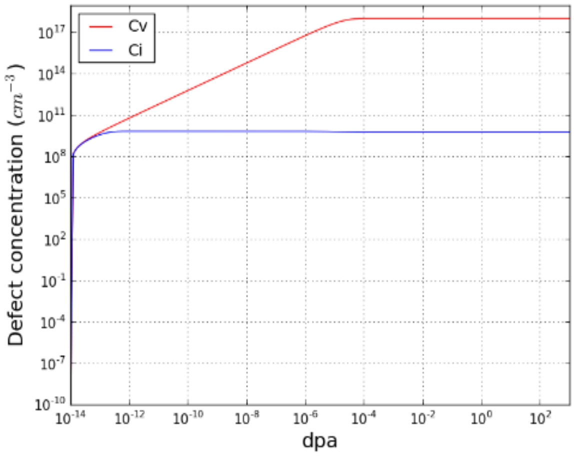

Fig. 3. Interstitial and vacancy concentration in aluminum matrix at 328 K.

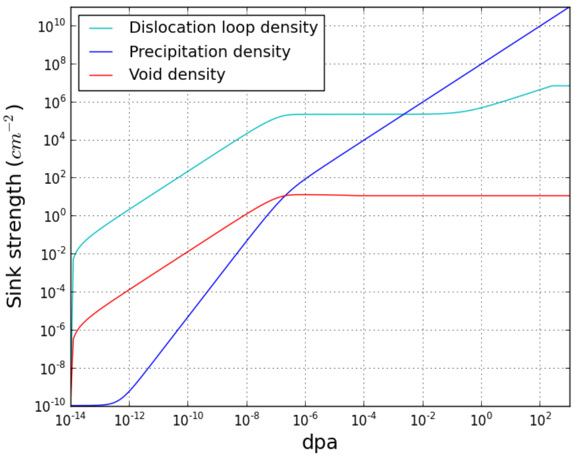

Fig. 4. Density change of sink in aluminum matrix at 328 K.

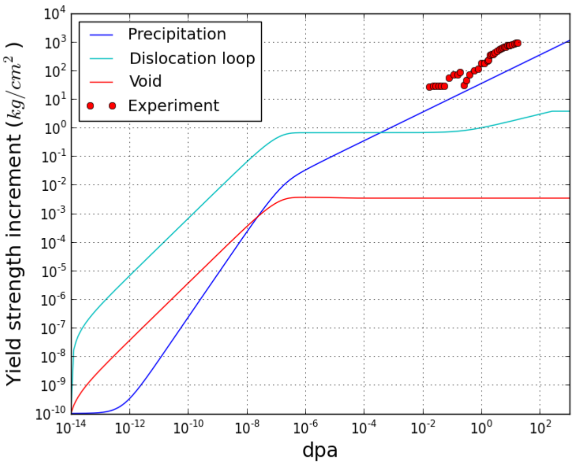

Fig. 5. Yield strength increment of aluminum at 328 K.

In Fig. 3, defect concentration in aluminum matrix shows typical high sink density behavior. Initially, defect concentrations are increasing and then both of interstitial and vacancy concentration are saturated certain amount. In this region, recombination effect can be neglected by high sink density. Therefore, interstitial concentration is not decreased when it compared with low sink density that.

In Fig. 4, change of sink density with dpa is revealed. Precipitate density shows linear increasing with dpa. However, density of dislocation loop and void is saturated at certain dpa region because defect flux become zero value.

In Fig. 5, increment of yield strength shows same tendency with sink density because irradiation hardening is proportional to density with sink. Therefore, precipitation effect on hardening was most dominant and dislocation loop and void can be neglect able. Result of experiment and calculation result in this study show a little disagreement.

4. Discussion

Radiation induced swelling and hardening are depend on three factors, sink density, defect concentration and bias factor. In this study, various simplified assumptions were adopted in these three factor for the simulation.

Firstly, among these three parameters, density of each sink was calculated by number density which is obtained from experiment result. It was assumed that sink will be saturated at certain dpa region. However, it is not clear that number density has proper value at any given dpa. Moreover, there is not in situ experiment equipment which could analyze change of sink density with dpa and temperature. Therefore, theoretical analysis of this sink is essential in research of MFRDT. However, there is little research about modeling of precipitation effect on other sinks in aluminum matrix.

Secondly, bias factors was assumed that interstitial has higher absorption rate batter than vacancy that because of sink strain field. However, this value has not been verified in aluminum matrix. In order to verify this value, literature study will be carry out about physical chemistry. And then this value will be verified in next research stage.

Lastly, defect concentration was obtained by very simplified model. In defect generation, it was assumed that cluster defects and mobile cluster also are neglected. Moreover, generated interstitial was assumed that interstitial dislocation is only sink which interstitial could be agglomerated while vacancy could generate only void in aluminum matrix. These assumption is over simplified for simulation. Therefore, these assumption should be revised by reflecting the actual defect reaction phenomena.

Recently, various research has been done to understand reaction mechanism of defect in matrix. However, in order to analyze this fundamental reaction mechanism, modeling in atomic scale should be established from nano to micro-scale. Although, MD simulation could explain behavior of atom in nano second, it is impossible to extend its capability to micro second. Therefore, another simulation tool should be suggested.

However, it is hard to simulate the reaction behavior by using deterministic method because all possible reaction mechanism should be calculated by theoretically before describe envious and various reaction module in detail. Therefore, probabilistic method could be one solution in this problem. Most recently, there was kMC modeling have been done about vacancy and helium cluster [6]. This model could be expend to precipitation effect on aluminum. Therefore, kMC modeling will be conducted in next research stage.

After kMC simulation carried out, defect rate equation of MFRDT will be modified because cluster effect on the defect generation term should be revised. From this work, irradiation

5. Conclusions

The aim of this study is understanding of radiation effect on sink behavior. From the simplified reaction mechanism, defect concentration, sink density and irradiation hardening are calculated at each sink type. Transmutation effect was mostly dominant and dislocation loop and void effect were negligible. From the KMC simulation, these extended defect behaviors will be examined in next research step.

6. Acknowledgement

This work was financially supported by the Korean Nuclear R&D program organized by the National Research Foundation (NRF) in support of the Ministry of Science, ICT and future Planning (MSIP).

Reference

[1] K. Farrell, Performance of aluminum in research Reactors, Comprehensive Nuclear Materials, 5, pp. 143-175 (2012)

[2] K. Farrell and R. T. King, Tensile properties of neutron-irradiated 6061 aluminum alloy in annealed and precipitation-hardened conditions, Effect of Radiation on Structural Materials, pp. 440-449 (1979)

[3] S.R. MacEwen and G.J.C. Carpenter, Calculations of irradiation growth in zirconium. Journal of Nuclear Materials, 90(1-3), pp. 108-132 (1980)

[4] Pedraza, A.J. and D. Fainstein-Pedraza, Irradiation growth of zirconium-base alloys: II. Annealed material. Journal of Nuclear Materials, 88(2–3): p. 236-248 (1980)

[5] C.H. Woo, Theory of Irradiation deformation in non-cubic metals effects of anisotropic diffusion. Journal of Nuclear Materials, 159: p. 237-256, (1988)

[6] M.J. Caturla, T. Diaz de la Rubia, M. Fluss, Radiation growth of HCP metals under cascade damage conditions, Journal of Nuclear Materials, 323, pp 163-168, (2003)