

## Irradiation growth modeling of polycrystalline zirconium and zircaloy for nuclear reactor

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### 1. Introduction

Neutron irradiation occur many materials degradation in nuclear reactor such as deformation, hardening and embrittlement. Among those phenomenon, material deformation is one of the most important phenomena because it is directly related with safety criteria. There are three representative module of radiation induced deformation. The first is irradiation growth which is volume conservative distortion of a solid without an applied stress, the second is swelling which is the isotropic volume expansion of a solid without any applied stress, the third is creep which is the volume conservative distortion by an applied stress. Zirconium, hafnium, titanium are representative materials which exhibit the irradiation growth. Among these materials, zirconium is widely used as a core material because of its excellent neutron interaction and corrosion resistance properties.

Hence, zirconium deformation behavior has been demonstrated by many experiments and theory since the 1960s [1-2]. Thanks to these efforts, it was revealed that the dislocation loop, dislocation line and grain boundary are major parameter in irradiation growth of zirconium. From this result, much theoretical work could be done based on rate theory [3-4]. These models predict successfully cold worked polycrystalline. However, these models failed to explain of annealed polycrystalline growth. At the end of the 1980s, Woo [5] change the traditional framework by using difference anisotropy diffusion (DAD) concept. In this framework, irradiation growth could be explained qualitatively in whole crystal systems such as a single crystal, as well as annealed and cold-worked polycrystalline samples. However, it was reveal that DAD could not explain quantitatively the high growth rate of cold worked zirconium at the high temperature region.

In 1993, Holt [6] demonstrated production bias modeling (PBM) effects on the zirconium. From this assumption, growth are well matched with irradiation growth modeling. However, unfortunately, quantitative modeling of annealed polycrystalline had not been done as opposed to the cold-worked polycrystalline since it was difficult to model the sink and texture effect on annealed polycrystalline compare with cold work.

### 2. Irradiation growth modeling

The origin of the irradiation growth is atom rearrangement by defect flux to sink. In 1979, Holt [7] quantitatively established defect flux to sink such as dislocation loop, dislocation line and grain boundary. In his papers, growth is calculated by sink orientation.

#### 2-1 Single Crystal growth equation

The growth strain along the a-axis of the single crystal was calculated. For the case of the single crystal, the dislocation loop is the only effective sink in the single crystal.

$$\frac{dG_a}{dt} = A_{lp} \times \rho_{lp} \times (Z_{lp}^i D_i C_i - Z_{lp}^v D_v C_v) \quad (1)$$

$A_{lp}$ ; Average strain factor of dislocation loop

$\rho_{lp}$ ; Grain boundary sink strength

$Z_{lp}^{i,v}$ ; Interstitial and Vacancy bias factor of dislocation loop in prism plane

#### 2-1 Polycrystalline Growth Equation:

Irradiation growth in a particular direction “d” was calculated from the defect rate equation. Anisotropy factor was deduced by using the Kearn factor. The grain boundary sink strength was divided by 2 as it is assumed that the prism plane and basal plane distributions are equal. Unlike single crystals, dislocation lines were also considered to contribute to the grain boundary sink effects. Growth strain along the a-axis was determined by the sink strength of the prism plane.

$$\begin{aligned} \frac{dG_d}{dt} = & A_{poly} \times [A_{dp} \times \rho_{dp} \times (Z_{dp}^i D_i C_i - Z_{dp}^v D_v C_v) \\ & + A_{lp} \times \rho_{lp} \times (Z_{lp}^i D_i C_i - Z_{lp}^v D_v C_v) \\ & + (k_{gb} / 2) \times (Z_{gb}^i D_i C_i - Z_{gb}^v D_v C_v)] \end{aligned} \quad (2)$$

$A_{poly}$ ; Anisotropy factor of polycrystal

$A_{dp}$ ; Average strain factor of dislocation line

$\rho_{dp}$ ; Dislocation line density in the prism plane

$k_{gb}/2$ ; Sink strength grain boundary

$Z_{dp}^{i,v}$  and  $Z_{gb}^{i,v}$ ; Interstitial and Vacancy bias factor of dislocation line and grain boundary in the prism plane

### 3. Defect rate equation for growth modeling

The PBM assumption is mobile point defect and immobile cluster defect are generated unequal number. Before the PBM concept, freely migrating point defect is only defect source of traditional rate equation. To reveal PBM effect on the growth, traditional defect rate equation is used to calculate annealed polycrystalline growth. In the traditional rate equation, sink effect are define by density cubic centimeter and DAD effect are added by capture efficiency factor ' $Z_{i,v}$ '. Other parameters such as defect generation, recombination and diffusion coefficient is quoted from Christien papers [8].

#### 3-1 framework of defect rate equation

To develop an defect rate equation, the major sink effect must be defined by each matrix system because flux of defects to a sink is the origin of the irradiation growth phenomenon. Defect flux to sink is proportional to sink density. The sink density function has been established by many research efforts. The dislocation loop is a time-dependent function and the grain boundary sink density is a function dependent on matrix geometry. Table 1 shows the major sink densities.

Table 1. Major sink density in cubic centimeter

| Sink Matrix             | Loop ( $cm^{-2}$ ) | Dislocation Line ( $cm^{-2}$ ) | GB( $cm^{-2}$ )   |
|-------------------------|--------------------|--------------------------------|-------------------|
| Single crystal          | Time dependent     | $1 \times 10^7$                | 0                 |
| Annealed polycrystal    | Time dependent     | $1 \times 10^7$                | $3.3 \times 10^8$ |
| Cold-worked polycrystal | Time dependent     | $8 \times 10^9$                | $2.4 \times 10^9$ |

#### 3-2 Traditional defect rate equation

Defect concentration was calculated by the defect rate equations which were solved with the Python program.

$$\frac{dC_v}{dt} = K_o - K_{iv}C_iC_v - Z_v\rho C_vD_v \quad (3)$$

$$\frac{dC_i}{dt} = K_o - K_{iv}C_iC_v - Z_i\rho C_iD_i \quad (4)$$

$K_o$ ; Defect production rate

$K_{iv}$ ; Vacancy-interstitial recombination

$\rho$ ; Total sink strength

$Z_i$  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 diffusion coefficient

### 4. Result

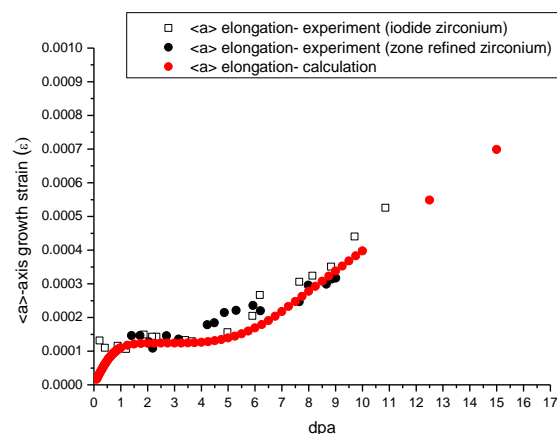


Fig. 1. Irradiation growth Strain of single crystal sample at 553 K.

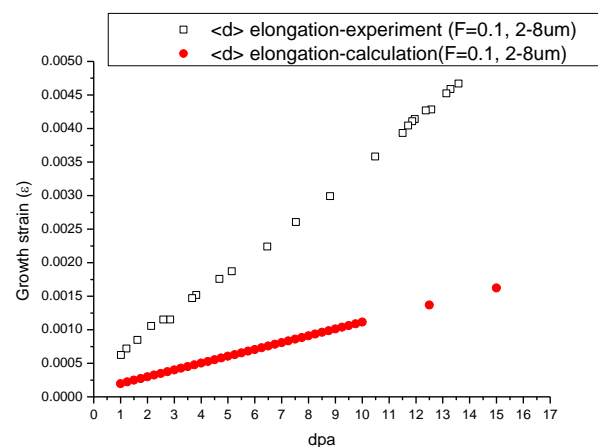


Fig. 2. Irradiation growth strain of cold-worked polycrystalline sample at 553 K.

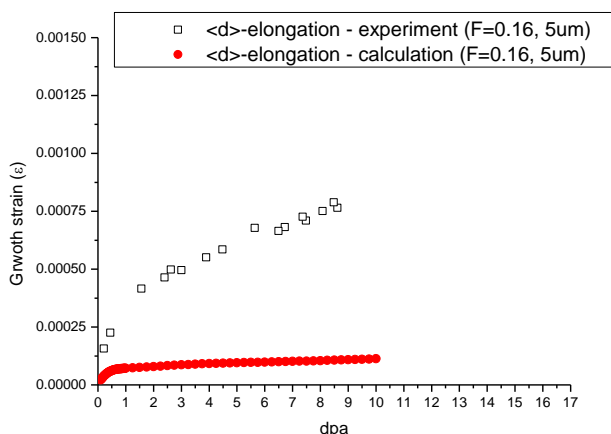


Fig. 3. Irradiation growth strain of annealed polycrystalline sample at 553 K.

## 5. Discussion

Irradiation growth is one of the most important phenomenon in the nuclear safety analysis. However there was not general irradiation growth modeling of polycrystalline for various temperature region. Therefore this work is very important for the nuclear safety analysis. However growth modeling is very difficult because it should be combined with defect rate equation. Both of defect rate equation and irradiation growth modeling are not easy to establish because, in order to realistic modeling, we should consider various parameter such as diffusion coefficient, sink strength, capture efficiency, and anisotropy factor. From these parameters, growth result could be modified

Historically defect rate equation are developed by many researchers. In 1993, Holt [6] explain PBM effects on the zirconium metal. From this assumption, experiment result are well matched with irradiation growth modeling form research reactor to commercial reactor temperature. PBM framework could explain discrepancy between modeling and experiment. However, in this paper, defect rate equation are not fully developed. Therefore, PBM will be studied next research stage. Here PBM are briefly reviewed.

### 5-1 framework of defect rate equation

In PBM frameworks, defect cluster are generated by different quantity because of different formation and migration energy. These defect cluster assume to be immobile however interstitial dislocation loop which is developed by cluster assume to be mobile. From these assumption, defect flux to sink should be defined by each sink.

### 5-2 PBM included defect rate equation

Defect concentration will be calculated by the defect rate equations which will be solved with the Python program.

$$\frac{dC_v}{dt} = K(1 - \varepsilon_v) - K_{iv}C_iC_v - Z_v\rho C_v D_v \quad (5)$$

$$\frac{dC_i}{dt} = K(1 - \varepsilon_i) - K_{iv}C_iC_v - Z_i\rho C_i D_i \quad (6)$$

$K_0$ ; Defect production rate  
 $\varepsilon_v$  and  $\varepsilon_i$ ; Vacancy Interstitial cluster fraction of total defect  
 $K_{iv}$ ; Vacancy and Interstitial recombination  
 $\rho$ ; Total sink strength  
 $Z_i$  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 diffusion coefficient

Despite of these tough works, many researcher are establish various modeling about irradiation phenomena by using appropriate assumption. Therefore, this work will be developed with same tendency. And then eventually, modeling would explain appropriately real irradiation growth phenomenon.

## 6. Summary

The traditional growth modeling are conducted and PBM mechanism for irradiation growth is reviewed. Traditional growth modeling result are well matched single crystal zirconium but polycrystalline did not.

1. Single crystal zirconium growth are well matched with experimental result. Detail analysis of sink strength and defect flux should be done next research stage.
2. In case of poly crystalline zirconium, modeling result are lower than experiment results. This result could be modified by adopting PBM framework or modifying diffusion coefficient

## 7. Acknowledgement

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## 8. Reference

- [1] A. Rogerson, Irradiation growth in zirconium and its alloys. Journal of Nuclear Materials, 159(C), pp. 43-61 (1988)
- [2] G.J.C. Carpenter, R.H. Zee, and A. Rogerson, Irradiation growth of zirconium single crystals: A review. Journal of Nuclear Materials, 159(C), pp. 86-100 (1988)
- [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] R.A. Holt and C.H. Woo, Production bias - a potential driving force for irradiation growth. Journal of Nuclear Materials, 205: p. 293-300 (1993)
- [7] R.A. Holt, and E.F. Ibrahim, Factors affecting the anisotropy of irradiation creep and growth of zirconium alloys. Acta Metallurgica, 27(8) pp. 1319-1328 (1979)
- [8] F. Christien, and A. Barbu, Cluster dynamics modelling of irradiation growth of zirconium single crystals. Journal of Nuclear Materials, 393(1): p. 153-161, (2009)