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# Modeling of neutron irradiation response for B2-type ordered alloys

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**Abstract**

B2 type ordered alloys with high symmetry have been considered as a possible attractive new material for application in fusion environments during severe irradiation field because of self-restoring phenomena during irradiation. Recently, a great deal of effort has been made to analyze the irradiation response by using multi-scale modeling. Our attention has been focused on utilization of the order–disorder transition. The irradiated state is associated with critical phenomena and can be expressed as two competing processes: ordering and disordering. A preliminary approach can be proposed to simply and quickly estimate the neutron-irradiated state of a B2 type ordered alloy as a function of temperature and displacement rate by using two measures of order for determining whether re-ordering under irradiation is preferred or not.
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## 1. Introduction

Radiation damage processes consist of various stages such as elastic collisions, defect aggregation and annihilation processes, nucleation and growth due to diffusion, and crack initiation and propagation. Numerous efforts have recently been made to clarify the irradiation response of materials through the use of multi-scale modeling. If the irradiated state can be easily described using a simple diagram that is a function of temperature, displacement rate, and displacement dose, we may obtain rough guidance or how to choose a target material for service in a severe irradiation environment without having to account for detailed effects such as cluster interactions and atom vibration.

Therefore, we have concentrated on using the traditional order–disorder transition that is described by the Ising model on a rigid ideal lattice with pair, but with otherwise arbitrary interatomic interactions. The irradiated state is associated with critical phenomena and can be expressed as two competing processes; one is ordering facilitated by defect migration and the other is disordering induced or accelerated by neutron irradiation. The degree of Bragg–Williams' long range order corresponds to composition fluctuations with a long wavelength. On the other hand, the degree of Warren–Cowley's short range order is correlated with cooperative phenomena due to the continuity of the lattice. In this paper, a preliminary approach is introduced to estimate simply and quickly the neutron-irradiated state of an ordered alloy as a function of temperature and displacement rate without taking into consideration the detailed microstructural evolution of the specimens. This is done by using two kinds of degrees of order for determining whether the re-ordering is the preferred process under irradiation or not.

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## 2. Order–disorder transition under irradiation

The competing processes of ordering and disordering take place under irradiation. In case of an ordered alloy,

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the degree of order can also be described by using the Bragg–Williams' long range order parameter or the Warren–Cowley's short range order parameter. The degree of long range order Bragg–Williams' long range order of a B2 type ordered alloy under irradiation can be defined by

$$S = \frac{C_A^{\alpha} - C_A}{1 - C_A} = \frac{C_B^{\beta} - C_B}{1 - C_B} \quad (0 \leqslant S \leqslant 1,\ C_A + C_B = 1), \tag{1}$$

where $\alpha$ and $\beta$ denote $\alpha$ sublattice and $\beta$ sublattices in an ordered structure, and the subscripts $A$ or $B$ indicate the $A$ atom or $B$ atom, respectively.

The total disordering rate can be expressed by [1–3]

$$\frac{dS}{dt} = \left[\frac{dS}{dt}\right]_{\text{disordering}} + \left[\frac{dS}{dt}\right]_{\text{ordering}} = -\varepsilon\phi S + K(1-S)^2. \tag{2}$$

The first term in the right hand part describes the disordering rate and the second term is the ordering rate.

Parameters used in the equation are as follows:

- $\varepsilon$ disordering efficiency (effective number of replacements per displacement)
- $\phi$ defect production rate (displacement rate: dpa/s)
- $K$ rate constant for the reordering as a function of temperature,

$$K = (Z_\alpha + Z_\beta - 2)C_v \cdot v \cdot C_A C_B \exp\left(-\frac{E}{kT}\right), \tag{3}$$

- $Z_\alpha$, $Z_\beta$ coordination number of $\alpha$ sublattice or $\beta$ sublattice
- $C_v$ vacancy concentration
- $v$ frequency factor
- $C_A$, $C_B$ atomic fraction of $A$ atoms and $B$ atoms
- $E$ vacancy migration enthalpy
- $k$ Boltzmann constant
- $T$ irradiation temperature.

The competition between the ordering process and disordering process leads to the following equation

$$\frac{dS}{dt} = -\varepsilon\phi\{S - R(1-S)^2\} = \varepsilon\phi R(S-\alpha)(S-\beta) = 0. \tag{4}$$

A solution of $S$ requires values of be $\beta$ in $\alpha$, $\beta$ which satisfy the condition of $0 \leqq S \leqq 1$.

$$\alpha, \beta = \frac{2R+1}{2R} \pm \left(\frac{4R+1}{4R^2}\right)^{\frac{1}{2}} \quad (\beta < \alpha,\ 0 \leqq \beta \leqq 1,\ 1 < \alpha), \tag{5}$$

where

$$R = \frac{K}{\varepsilon\phi} = \frac{S_{\text{eq}}}{(1 - S_{\text{eq}})^2} \quad (0 \leqq R < \infty)\ [3,4]. \tag{6}$$

$S_{\text{eq}}$ is the steady-state degree of order, which is achieved at long irradiation times.

Therefore, the following solution can be written as

$$\begin{aligned} S &= \beta + \frac{(\alpha-\beta)(1-\beta)}{(\alpha-1)\exp\{\varepsilon R(\alpha-\beta)\phi t\} + (1-\beta)} \\ &= 1 + \frac{1}{2R}\left\{1 - (4R+1)^{\frac{1}{2}}\right\} \\ &\quad + \frac{4(4R+1)^{\frac{1}{2}}}{\left\{1 + (4R+1)^{\frac{1}{2}}\right\}^2 \exp\left\{(4R+1)^{\frac{1}{2}}\varepsilon\phi t\right\} + 4R} \\ &= 1 + \frac{(\varepsilon\phi)^{\frac{1}{2}}}{2K}\left\{(\varepsilon\phi)^{\frac{1}{2}} - (4K+\varepsilon\phi)^{\frac{1}{2}}\right\} \\ &\quad + \frac{4(\varepsilon\phi)^{\frac{1}{2}}(4K+\varepsilon\phi)^{\frac{1}{2}}}{\left\{(\varepsilon\phi)^{\frac{1}{2}} + (4K+\varepsilon\phi)^{\frac{1}{2}}\right\}^2 \exp\left\{(\varepsilon\phi)^{\frac{1}{2}}(4K+\varepsilon\phi)^{\frac{1}{2}}t\right\} + 4K}. \end{aligned} \tag{7}$$

Fig. 1 shows the temperature dependence of the irradiated states of a B2 type ordered alloy calculated using Eq. (7). It has been shown that the degree of long range order at 370 K, which was nearly zero, increased at 520 K due to the restoring phenomena that took place in the alloy. These phenomena have already been reported for a Ti–Ni alloy with the B2 type ordered structure under neutron irradiation in Japan Materials Testing Reactor (JMTR) of JAERI [5].

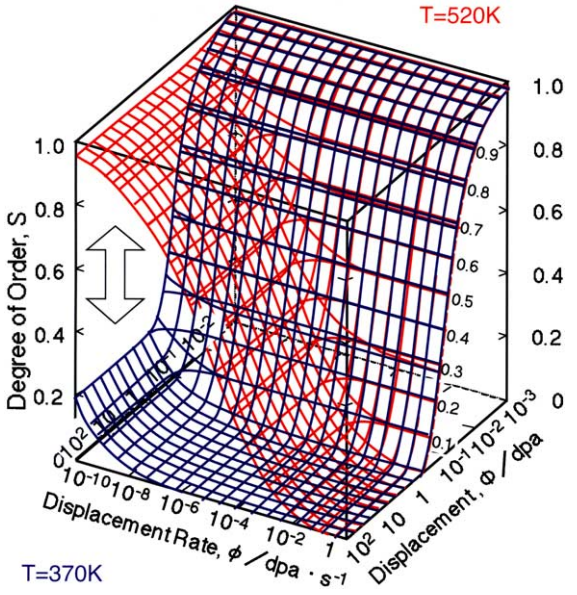


Fig. 1. Temperature dependence of irradiated states of a B2 type ordered alloy.

The relationships between the Bragg–Williams' long range order parameter and Warren–Cowley's short range order parameter $\sigma$, which reveals how inhomogeneous the damage is, can be obtained as follows.

The number of $A$–$A$ atomic pairs in a binary alloy can be written as [6]

$$N_{AA} = \frac{1}{2} N \cdot Z \cdot C_A \cdot p_{AA}, \tag{8}$$

where $p_{AA}$ is the probability of an $A$ atom to have a nearest neighbor $A$ atom, and $Z$ is the coordination number.

The probability for a site of the $\alpha$ sublattice or $\beta$ sublattice to be occupied by an $A$ atom is defined by $p_A^i (i \in \alpha)$, $p_A^j (j \in \beta)$, respectively:

$$p_{AA} = \frac{\frac{1}{2}N}{\frac{1}{2}N \cdot C_A \cdot Z} \sum_{i \neq j} p_A^i p_A^j = \frac{1}{Z \cdot C_A} \sum_{i \neq j} p_A^i p_A^j \quad (i \in \alpha,\ j \in \beta). \tag{9}$$

On the other hand, the long range order parameter $S$ and $p_A^\alpha$ is expressed by

$$S = \frac{p_A^\alpha - C_A}{1 - v} = \frac{p_A^\alpha - C_A}{\gamma}, \tag{10}$$

from the relation between $C_A$ and $v$ (the relative concentration of $\alpha$ sublattice sites), where $\gamma$ is obtained by Ref. [6]

$$C_A \leqslant v, \quad \gamma = \frac{C_A(1-v)}{v}, \quad C_A \geqslant v, \quad \gamma = 1 - C_A. \tag{11}$$

From these relationships, $p_A^\alpha = S \cdot \gamma + C_A$.

In a B2 type ordered alloy, which has the condition of $Z_\alpha = Z_\beta = Z$,

$$\begin{aligned} p_{AA} &= \frac{1}{ZC_A}[\{C_A Z_\alpha + (1 - C_A) Z_\beta\}(C_A + \gamma \cdot S) \\ &\quad \times (1 - C_A - \gamma \cdot S)] \\ &= C_A - \frac{\gamma^2 S^2}{C_A} \end{aligned} \tag{12}$$

can be obtained [7].

The short range order parameter of $\sigma_1$ can be defined by

$$\sigma_1 = 1 - \frac{p_{AB}}{C_B} = 1 - \frac{1 - p_{AA}}{C_B} \quad \left\{ \begin{array}{c} p_{AA} + p_{AB} = 1 \\ p_{BA} + p_{BB} = 1 \\ C_A p_{AB} = C_B p_{BA} \end{array} \right\}, \tag{13}$$

where the short range order of $\sigma$ (the first nearest neighbor) can be obtained by using the long range order of $S$

$$\sigma_1 = -\frac{\gamma^2 S^2}{C_A(1 - C_A)}. \tag{14}$$

## 3. Irradiated state diagram (ISD) of a B2 type ordered alloy

The degree of long range order $S$ is gradually decreased with increasing damage from $S = 1$ (before irradiation), until a steady-state value is reached under irradiation.

In Eq. (7), the first term of $\beta$ corresponds to the degree of long range order at steady state ($= S_{\text{eq}}$). The second term depends on irradiation time, and reveals the time dependence and approaches the steady-state degree of order with increasing irradiation time.

$$\begin{aligned} S_{\text{th1}} &= 1 + \frac{1}{2R}\left\{1 - \frac{(4R+1)^2}{(2R+1)(4R+1) + 4R^2}\right\} \\ &= 1 + \frac{\varepsilon\phi}{2K}\left\{1 - \frac{(4K + \varepsilon\phi)^2}{(2K + \varepsilon\phi) \cdot (4K + \varepsilon\phi) + 4K^2}\right\}, \end{aligned} \tag{15}$$

$$S_{\text{th2}} = \frac{2R}{2R+1} = \frac{2K}{2K + \varepsilon\phi} \tag{16}$$

for $0 \leqq S_{\text{eq}} < S_{\text{th2}} < S_{\text{th1}} < 1$.

The relationships among the first threshold value of $S_{\text{th1}}$ at which long range order of $S$ is drastically reduced under irradiation, the second threshold value of $S_{\text{th2}}$ attained at an almost steady-state value after decreasing $S$, and the parameter of $R$, can be obtained by using the second and first partial differential coefficients of a continuum function of $R$ with respect to $R$ (temperature or displacement rate).

The steady-state degree of order can be obtained by

$$\begin{aligned} S_{\text{eq}} &= 1 + \frac{1}{2R}\left\{1 - (4R+1)^{\frac{1}{2}}\right\} \\ &= \frac{1}{2K}\left[2K + \varepsilon\phi - \{\varepsilon\phi(4K + \varepsilon\phi)\}^{\frac{1}{2}}\right]. \end{aligned} \tag{17}$$

Furthermore, the relationship between irradiation temperature $T$ and displacement rate $\phi$ can be written as follows:

$$-\frac{E}{kT} - \frac{1}{2}\ln\phi = \ln R + \ln\frac{\varepsilon}{(Z_\alpha + Z_\beta - 2) \cdot (v/z)^{\frac{1}{2}} \cdot C_A C_B}. \tag{18}$$

In the case long range order reaches steady state, the relationship between displacement rate and irradiation temperature can be expressed by

$$\begin{aligned} \ln\phi_{\text{eq}} &= -\frac{2E}{kT_{\text{eq}}} + 2\ln\frac{(Z_\alpha + Z_\beta - 2) \cdot (v/z)^{1/2} \cdot C_A C_B}{\varepsilon} \\ &\quad - 2\ln\frac{S_{\text{eq}}}{(1 - S_{\text{eq}})^2}, \end{aligned} \tag{19}$$

where $T_{eq}$ is a temperature at which the degree of long range order is attained at steady state.

$$\ln \phi_{th2} = -\frac{2E}{kT_{th2}} + 2\ln\frac{(Z_\alpha + Z_\beta - 2)\cdot(v/z)^{1/2}\cdot C_A C_B}{\varepsilon} - 2\ln\frac{S_{th2}}{2(1-S_{th2})}, \quad (20)$$

$$\ln \phi_{th1} = -\frac{2E}{kT_{th1}} + 2\ln\frac{(Z_\alpha + Z_\beta - 2)\cdot(v/z)^{1/2}\cdot C_A C_B}{\varepsilon} - 2\ln\frac{(4-3S_{th1}^2)^{\frac{1}{2}} + 3S_{th1} - 2}{12(1-S_{th1})}. \quad (21)$$

$T_{th2}$, $T_{th1}$, $\phi_{th2}$ and $\phi_{th1}$ are the first threshold temperature at which the long range order abruptly decreased, the second threshold temperature at which long range order attained near steady state, and the first and second threshold displacement rates.

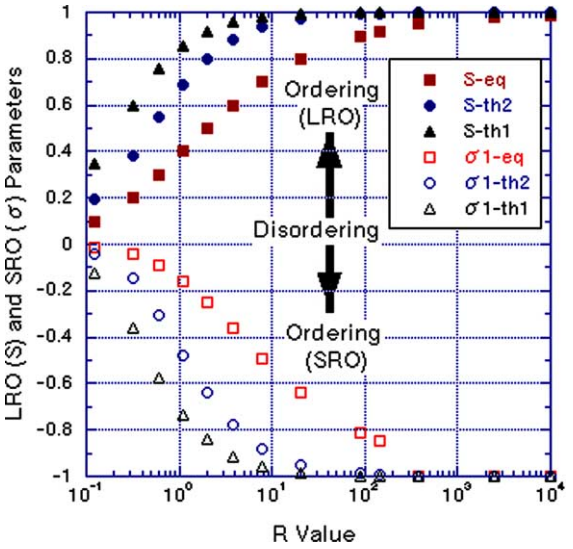


Fig. 2. LRO parameter ($S$) and SRO parameter ($\sigma$) as a function of $R$ value for irradiated states.

## 4. Results and discussion

The degree of Bragg–Williams' long range order corresponds to the composition fluctuation long wavelength. On the other hand, the degree of Warren–Cowley's short range order is correlated with cooperative phenomena due to the continuity of lattice. Table 1 shows the long range order (LRO) parameter $S$ and the short range order (SRO) parameter $\sigma$ for irradiated states obtained by Eqs. (14)–(17). The LRO parameter $S$ and SRO parameter $\sigma$ are shown in Fig. 2 as a function of the $R$ value. If a disordering process is the predominant factor in irradiated states for increasing displacement damage, the LRO and SRO parameters approach nearly to zero. On the other hand, if the LRO parameters go almost to unity and SRO parameters reach −1, this indicates the ordering of unlike atoms. In some cases, the SRO parameter indicates the possibility of clustering (the ordering on like atoms), if ordering is the predominant process rather than disordering process.

Fig. 3 shows the various displacement rate and temperature regime of an irradiated state diagrams (ISD) for a B2 type ordered alloy. In the figure, changes

Table 1
LRO parameters ($S$) and SRO parameters ($\sigma$) for irradiated states

| $R$ value | Bragg–Williams' long range order parameter $S$ | | | Warren–Cowley's short range order parameter $\sigma_1$ (first nearest neighbor) | | |
|---|---|---|---|---|---|---|
| | Steady state | Threshold value | | Steady state | Threshold value | |
| | $S_{eq}$ | $S_{th2}$ | $S_{th1}$ | $\sigma_{1-eq}$ | $\sigma_{1-th2}$ | $\sigma_{1-th1}$ |
| 0.12 | 0.1 | 0.2 | 0.35 | −0.01 | −0.04 | −0.12 |
| 0.31 | 0.2 | 0.39 | 0.6 | −0.04 | −0.15 | −0.36 |
| 0.61 | 0.3 | 0.55 | 0.76 | −0.09 | −0.3 | −0.57 |
| 1.1 | 0.4 | 0.7 | 0.86 | −0.16 | −0.48 | −0.73 |
| 2 | 0.5 | 0.8 | 0.92 | −0.25 | −0.64 | −0.84 |
| 3.8 | 0.6 | 0.88 | 0.96 | −0.36 | −0.78 | −0.91 |
| 7.8 | 0.7 | 0.94 | 0.98 | −0.49 | −0.88 | −0.96 |
| 20 | 0.8 | 0.98 | 0.99 | −0.64 | −0.95 | −0.98 |
| 90 | 0.9 | 0.99 | *1.0 | −0.81 | −0.99 | *−1.0 |
| 148 | 0.92 | *1.0 | *1.0 | −0.85 | −0.99 | *−1.0 |
| 330 | 0.95 | *1.0 | *1.0 | *−1.0 | *−1.0 | *−1.0 |
| 2450 | 0.98 | *1.0 | *1.0 | *−1.0 | *−1.0 | *−1.0 |
| 9900 | 0.99 | *1.0 | *1.0 | *−1.0 | *−1.0 | *−1.0 |

* Denotes fully ordered states.

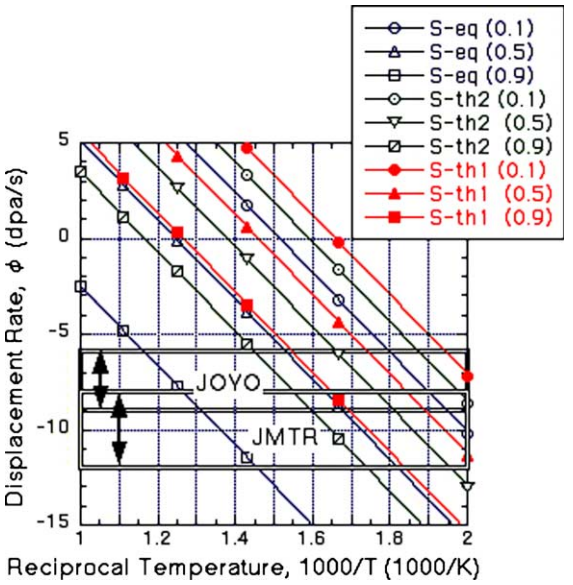


Fig. 3. Irradiated state diagrams (ISD) of displacement rate versus temperature for a B2 type ordered alloy.

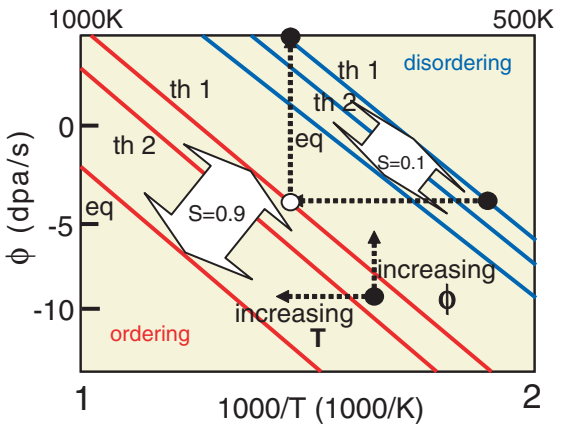


Fig. 4. Irradiated state diagram (ISD) of a B2 type ordered alloy.

in degrees of long range order at 0.1, 0.3 and 0.9 can be expressed as several kinds of lines in which LRO decreases abruptly ($S_{th1}$), decreases gradually and approaches a steady state ($S_{th2}$), and the steady-state value ($S_{eq}$). Irradiation conditions of the Experimental Fast Reactor of JOYO in JNC ($10^{-6}$ to $10^{-9}$ dpa/s) and the Japan Materials Testing Reactors of JMTR ($10^{-8}$ to $10^{-12}$ dpa/s) in JAERI are also indicated in the figure. The behavior of irradiated Ti–Ni alloys, which was obtained by electrical measurements, can be explained by this diagram [5].

If the irradiated state diagram (ISD) can be used for surveying the conditions of practical use for newly developed materials, the impact of a severe irradiation field can be easily and quickly obtained without having to account for the influence of complicated irradiation parameters into consideration. By comparing standard behavior with the degree of LRO obtained for the in-reactor conditions shown in Fig. 4, changes in the degree of order and material stability can be predicted under irradiation.

## 5. Conclusions

An order–disorder transition model under irradiation has been newly proposed for the B2 type ordered structure. The irradiated state can be expressed as two competing processes; one is ordering facilitated by defect migration and the other is disordering induced or accelerated by neutron irradiation. The preliminary approach indicates that the irradiated state diagram (ISD) can be introduced to simply and quickly estimate the neutron-irradiated state of a B2 type ordered alloy as a function of temperature and displacement rate. Analysis with two different degrees of order can be used to determine whether ordering or disordering will be the predominant factor governing the irradiated state.

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