

DIFFUSION AND AGGLOMERATION OF HELIUM IN STAINLESS STEEL

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ABSTRACT

Diffusion of helium and formation of helium bubbles in stainless steel in condition of atomic displacement are studied theoretically using standard rate equations [1]. Besides this bubble coalescence is assumed to result when collisions occur between bubbles as they migrate by surface diffusion through the solid [2]. The dissociative mechanism via self-interstitial *He* replacement is assumed to control helium diffusion and bubble formation. The theoretically analysis is based on the diatomic nucleation model where two helium atoms are assumed to compose a stable nucleus. For nucleation of interstitial loops, two atoms are assumed to compose a stable nucleus. In the present work is assumed that coalescence follows bubble collisions resulting from random migration of bubbles in Chandrasekhar's approximation.

INTRODUCTION

The introduction of helium into metals by ion implantation and nuclear transmutation can result in a serious loss of ductility, which is of great concern for performance of metals in nuclear power plants, high-flux spallation neutron sources and tritium storage.

In the present work, we study a model for helium diffusion and bubble formation in stainless steel implanted with energetic helium ions in the intermediate temperature range. The temperature dependence of helium diffusivity and bubble structures is our main concern. The model is simplified by ignoring the effects of grain boundaries. The migration and coalescence of bubbles is not ignored. While formation of bubbles at dislocations has to be considered in the model.

MODEL DESCRIPTION AND RATE EQUATIONS

The theoretical analysis is based on the diatomic nucleation model where two helium atoms are assumed to compose a stable nucleus. In this case bubble formation is controlled by the diffusion of helium. The usual vacancy mechanism is not likely operative because of the immobility of vacancies at ambient temperature. In the model we consider another possible mechanism for radiation-enhanced helium diffusion, the so-called "dissociative mechanism" by which *He* atoms dissociate from the vacancies to interstitial lattice sites as a result of recombination of Frenkel pairs. The dissociative mechanism via the self-interstitial/*He* replacement is expected to be operative in the intermediate temperature range, since self-interstitials and *He* atoms at interstitial sites have considerable mobility even at temperatures far below room temperature. It is suggested that mobile only complex containing two *He*-atoms (surface diffusion mechanism). Equilibrium complex of two or four *He*-atoms is pore.

In the following rate equations, kinetic evolution of self-interstitials, vacancies, interstitial loops and *He*-vacancy clusters (bubbles) are described according to [1]. Formation of bubbles on dislocations is described according to [3]. The meaning and values of the symbols, parameters and constants (in stainless steel) are described in Table 1.

For point defects and interstitial loops,

$$\begin{aligned} \frac{dC_i}{dt} = & K(1 - v_s C_i - v_s C_v) / \Omega - 4\pi r_i (D_i + D_v) C_i C_v - Z_i D_i C_i (\rho_d + \rho_l) - \\ & - 4\pi r_i^s D_i C_i C_{He}^s - 4\pi Z_i^b D_i C_i (\bar{r}_b C_b + \bar{r}_b^d C_b^d) - 8\pi r_i D_i C_i^2 \end{aligned} \quad (1)$$

$$\begin{aligned} \frac{dC_v}{dt} = & K(1 - v_s C_i - v_s C_v) / \Omega - 4\pi r_r (D_i + D_v) C_i C_v - Z_v D_v C_v (\rho_d + \rho_l) - \\ & - 4\pi r_{He}^v (D_v + D_{He}^i) C_v C_{He}^i - 4\pi Z_v^b D_v C_v (\bar{r}_b C_b + \bar{r}_b^d C_b^d) - 8\pi r_i D_i C_i^2 \end{aligned} \quad (2)$$

Table 1. Symbols, parameters and constants used in the calculation

Symbols	Meaning	Values used in calculation	Unit
r_r	Radius of recombination of Frenkel pairs	3.5×10^{-10}	m
r_i	Trapping radius between self-interstitials	3.5×10^{-10}	m
r_{He}^i	Trapping radius between interstitial <i>He</i> atoms	3.5×10^{-10}	m
r_i^s	Trapping radius between a self-interstitial and a substitutional <i>He</i> atom	3.5×10^{-10}	m
r_{He}^v	Trapping radius between a vacancy and a interstitial <i>He</i> atom	3.5×10^{-10}	m
r_{He}^s	Trapping radius between a interstitial <i>He</i> atom and a substitutional <i>He</i> atom	3.5×10^{-10}	m
D_i	Diffusivity of self-interstitials	$10^{-5} \exp(-E_i^m / k_b T)$	m ² /s
D_v	Diffusivity of vacancies	$5 \times 10^{-5} \exp(-E_v^m / k_b T)$	m ² /s
D_{He}^i	Diffusivity of interstitial <i>He</i> atoms	$\approx D_i$	m ² /s
D_{He}^d	Diffusivity of helium on dislocations	$\approx D_{He}^{eff}$	m ² /s
E_i^m	Energy of migration of interstitials	$0.15 \times 1.6 \times 10^{-19}$	eV
E_v^m	Energy of migration of vacancies	1.3	eV
$\bar{n}_{He}$	Average number of <i>He</i> within a bubble		
$\bar{n}_v$	Average number of vacancy within a bubble		
ρ_d	Density of pre-implantation dislocations	10^{11}	m ⁻²
ρ_l	Density of interstitial loops	$2\pi r_1 C_1$	m ⁻²
Z_i	Dislocation bias factor for interstitials	1.1	
Z_v	Dislocation bias factor for vacancies	1	
Z_{He}^i	Dislocation bias factor for helium	1.1	
Z_i^b	Bubble bias factor for interstitials	$\exp(-(P_{He} - 2\gamma / \bar{r}_b) \Omega / k_b T)$	
Z_v^b	Bubble bias factor for vacancies	1	
Z_{He}^b	Bubble bias factor for <i>He</i> atoms	1	
G_{He}	Generation rate of helium in matrix	$10^{-2}, 10^{-1}, 10^0$	appm/s
G_{He}^d	Generation rate of helium on dislocations		s ⁻¹
K	Atomic displacement rate	$10^{-6}, 10^{-5}, 10^{-4}$	dpa/s
a	Lattice constant	3.5×10^{-10}	m
b	Burger's vector	2.5×10^{-10}	m
b_v	Van der Waal's constant of helium	1.59×10^{-29}	m ³
Ω	Metal atomic volume	1.2×10^{-29}	m ³

v_s	Spontaneous recombination volume	100Ω	m ³
γ	Surface energy	2.0	J/m
μ	Shear module	100	GPa
k_b	Boltzmann's constant	1.38×10^{-23}	J/K
T	Temperature	300-900	K
D_s	Surface's diffusivity of Fe-atoms	$0.58 \exp(-Q/RT)$	m ² /s
Q	Energy activation	142	kJ/mol
R	Gas const.	8.31	J/mol

$$\begin{aligned} \frac{dC_{He}^i}{dt} = & G_{He} / \Omega + 4\pi r_r D_i C_i C_{He}^s - Z_{He}^d D_{He}^i C_{He}^i (\rho_d + \rho_l) - 4\pi r_{He}^v (D_v + D_{He}^i) C_v C_{He}^i - \\ & - 4\pi Z_{He}^b D_{He}^i C_{He}^i (\bar{r}_b C_b + \bar{r}_b^d C_b^d) - 4\pi r_{He}^s D_{He}^i C_{He}^i C_{He}^s - 8\pi r_{He}^i D_{He}^i C_{He}^{i2} \end{aligned} \quad (3)$$

$$\frac{dC_{He}^s}{dt} = 4\pi r_{He}^v (D_v + D_{He}^i) C_v C_{He}^i - 4\pi r_i^s D_i C_i C_{He}^s - 4\pi r_{He}^s D_{He}^i C_{He}^i C_{He}^s \quad (4)$$

$$\frac{dC_l}{dt} = 8\pi r_i D_i C_i^2 \quad (5)$$

$$\frac{d\bar{r}_l}{dt} = (Z_i D_i C_i - Z_v D_v C_v) b^{-1} \Omega \quad (6)$$

where $C_i, C_v, C_l, C_{He}^i, C_{He}^s$ are concentration of self-interstitials, vacancies, interstitial loops. He atoms at interstitial and substitutional sites, respectively. $\bar{r}_l$ is the mean radius of interstitial loops. Spontaneous recombination of Frenkel pairs is included in the first terms in Eqs. (1) and (2). The fourth term in Eq. (1) and second term in Eq. (4) represent the self-interstitial/ He replacement mechanism.

For formation of bubbles in the matrix,

$$\frac{dC_{4j}}{dt} = 8\pi D_2 r_{2j} C_{2j}^2, \quad (7)$$

$$\frac{dC_{2j}}{dt} = 4\pi \cdot r_l \cdot D_j C_j (C_j + C_s) - 2 \cdot \frac{dC_b}{dt} - J_{2j}, \quad (8)$$

$$J_{2j} = 4\pi D_2 (r_b + r_{2j}) \cdot C_{2j}, \quad (9)$$

$$D_2 = 3/2 \cdot \Omega^{4/3} D_s / (\pi r_{2j}^4), \quad (10)$$

where C_b is the number density of bubbles in matrix, C_{nj} - concentration of complex of n He -atom, r_{nj} - equilibrium rad. of complex, D_2 - dif. coeff. for surface diff. Migration [2], D_s - surface diffusion coefficient.

$$\frac{d\bar{n}_j}{dt} = 4\pi Z_{He}^b \bar{r}_b D_{He}^i C_j (C_j + C_s) + 2J_{2j}, \quad (11)$$

$$\frac{d\bar{n}_v}{dt} = 4\pi Z_v^b \bar{r}_b (D_v C_v - Z_i D_i C_i) + n_{v2} \cdot J_{2j}, \quad (12)$$

where $\bar{n}_j, \bar{n}_v$ are the average number of He atoms and vacancies, respectively contained in a

bubble, n_{v2} – number vacancies in two *He*-atoms complex. The growth of bubbles is dependent on the inner gas pressure P_{He} , which is described by Van der Waal's gas law

$$P_{He} = \bar{n}_{He} k_b T \left(\frac{3}{4} \pi \bar{r}_b^3 - \bar{n}_{He} b_v \right). \quad (13)$$

Bubbles are expected to grow by absorbing vacancies (i.e., bias-driven growth) when there is a considerable concentration of freely migrating vacancies, while they are expected to grow by pushing out some matrix atom planes when the inner gas pressure exceeds the critical value of mechanical stability $(P_{He} - 2\gamma / \bar{r}_b) \geq \mu b / \bar{r}_b$ (μ is the shear module of matrix),

$$(d\bar{r}_b)_{LP} = b / 4. \quad (14)$$

For nucleation of bubbles at dislocations, dislocations produced before *He* ion implantation (for example by cold working) or during implantation (by production of dislocation loops) are expected to supply equally favorable sites for bubble nucleation. The former are constant in density (denoted as ρ_d) while the latter increase (denoted as ρ_l) during *He* ion implantation.

$$\frac{dC_1}{dt} = f_z \cdot J_d - 8D_l C_1^2 / l_s^2 - 16D_l C_1 / l_s, \quad (15)$$

$$\frac{dC_2}{dt} = 8D_l C_1^2 / l_s, \quad (16)$$

$$\frac{d\bar{n}}{dj} = 8D_l C_1 / l_s + 4\pi \cdot r_b D_j C_j, \quad (17)$$

where C_1 , C_2 – liner's concentrations (m^{-1}) of *He*-atoms and bubbles on dislocation, n_j - the average number of *He* atoms, contained in a bubble, n_v - the average number of vacancies give equation (12),

$$J_d = Z_j D_j C_j, \quad (18)$$

$$D_l = C_j / (C_j + C_s) D_j \text{ (eff.diff.coeff.)}, \quad (19)$$

$$f_z = \Omega / (\rho_d + \rho_l) / b \text{ (corr.factor)}, \quad (20)$$

$$l_s = (1 - 2r_b C_2) / C_2 \text{ (distance between pore's surface)}. \quad (21)$$

RESULTS AND DISCUSSION

The time dependence of concentrations and number of vacancies and *He*-atoms in pore is shown in fig.1-4 ($T=873$ K, $K=10^{-6}$ apa/s, $K_{He}=1$ appm/s). The correct analysis in pore's nucleation on dislocation show significant differences with results [1].

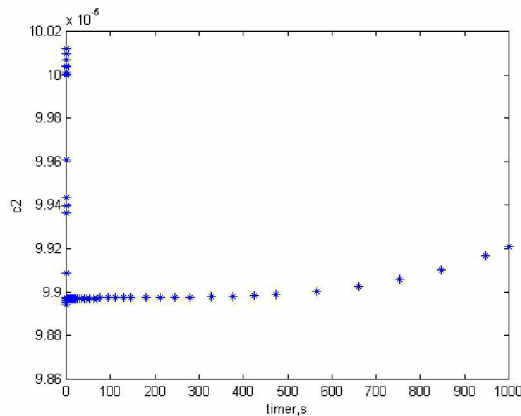


Fig.1. Line concentration of pore on dislocation versus time.

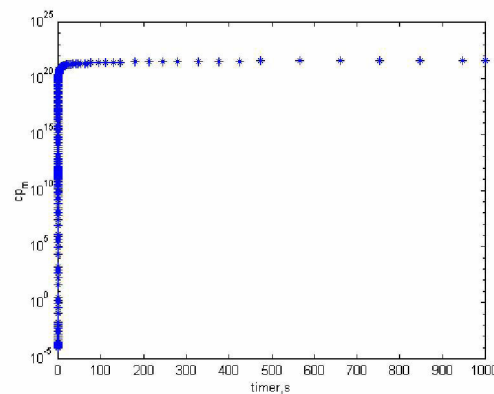


Fig.2. Volume concentration of pore in matrix versus time.

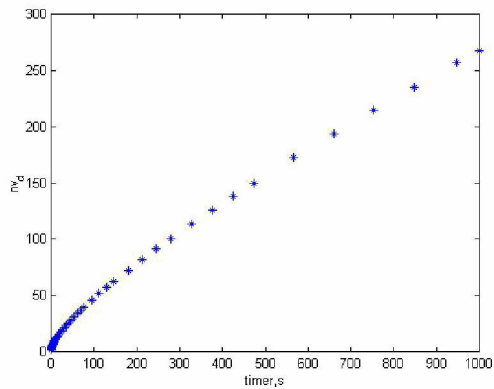


Fig.3. Number vacancies in pore on dislocation versus time.

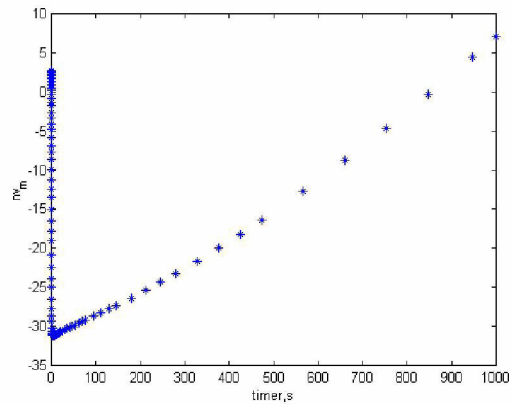


Fig.4. Number vacancies in pore in matrix versus time.

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