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Reaction kinetic analysis of reactor surveillance data

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ABSTRACT

In the reactor pressure vessel surveillance data of a European-type pressurized water reactor (low-Cu steel), it was found that the concentration of matrix defects was very high, and a large number of precipitates existed. In this study, defect structure evolution obtained from surveillance data was simulated by reaction kinetic analysis using 15 rate equations. The saturation of precipitation and the growth of loops were simulated, but it was not possible to explain the increase in DBTT on the basis of the defect structures. The sub-grain boundary segregation of solutes was discussed for the origin of the DBTT increase.

1. Introduction

The number of nuclear power reactors that have been in operation for more than 40 years is increasing. Neutron irradiation embrittlement of reactor pressure vessel steels (low-alloy steels) is the principal aging issue. Some surveillance data from these aged reactors indicate higher ductile brittle transition temperatures (DBTT) than those predicted by embrittlement correlation methods [1,2]. In particular, the precipitates are the main defect clusters, and only few loops were observed by transmission electron microscopy. No evidence of vacancy clusters was detected [3–5]. These are relatively high-Cu steels. The defect structure evolution obtained from surveillance data from the Genkai Unit 1 (0.12 wt %Cu steel) [4,5] was analyzed using reaction kinetic analysis [6]. The defect structure evolution of surveillance test pieces could be correctly reproduced by simulations.

Recently, surveillance test pieces of low-Cu steel (Mn:1.30, Ni:0.73, Mo:0.50, Si:0.23, Cr:0.11, C:0.022, S:0.012, P:0.011, Cu:0.04, V:0.01 wt%) from a European type pressurized water reactor (0.018, 0.045, 0.11 and 0.16 dpa) were tested. In addition to Mn-Ni-Si precipitates, many small dislocation loops were observed [7]. The loop density increased remarkably above 0.11 dpa. The volume fraction of Mn-Ni-Si clusters was saturated by the damage at 0.11 dpa, although isolated Mn and Ni remained in the matrix. This suggests the existence of a rate limiting factor for precipitation.

Reaction kinetic analysis using rate equations is one of powerful techniques to study damage structure evolution. Most of previous studies have been performed to obtain physical values such as migration energies of point defects (for example [8–10]). While some attempts have been made to simulate damage evolution by fitting the experimental results to clarify physical mechanisms underlying irradiation effects [11–14].

In this paper, the defect structure evolution obtained from surveillance data of the European type pressurized water reactor was simulated with an improved code adopted in Ref. [6]. We adjusted the coefficients of the rate equations to reproduce the surveillance data, considering effects due to the alloy element. To introduce the rate limiting factor for precipitation, the composition of precipitates was assumed to be G-phase like ($Mn_6Ni_{16}Si_7$) [15]. After the depletion of Si, the precipitation comes to an end, because Si was the minor element of the three constituents (Mn, Ni, Si) in the alloy.

2. Method

The model used for the calculations was also used in previous papers [6,13,14], and is based on the rate theory. It describes the reaction rates among point defects and their defect clusters. The following assumptions were made in the calculation:

(1) Two types of solutes, solute(G) which moves with interstitials and solute(M) which moves with vacancies are considered.

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(2) Interstitial-type dislocation loops are nucleation sites of precipitates.
(3) For the restriction of solutes to the precipitate, the composition of precipitates is assumed to be G-phase like ($Mn_6Ni_{16}Si_7$) [15]. After the depletion of Si, the precipitate cannot absorb any more solutes.
(4) Tri-interstitials and tri-vacancies are set for stable nuclei belonging to loops and voids, respectively.
(5) Mobile defects are interstitials, di-interstitials, solute(G)-interstitial pairs, vacancies, di-vacancies, and solute(M)-vacancy pairs.
(6) Loops + solute(M)-vacancy pair and loops + solute(G)-interstitial pairs are set for stable nuclei of solute-point defect clusters.
(7) The thermal dissociation of vacancies and interstitials is considered for di-vacancies, solute(M)-vacancy pairs, voids, and solute(G)-interstitial pairs.

The time dependence of 15 variables is calculated from 0 to approximately 1 dpa for the following quantities: the concentration of interstitials (C_I), di-interstitials (C_{2I}), interstitial clusters (interstitial-type dislocation loops, C_{IC}), vacancies (C_V), di-vacancies (C_{2V}), vacancy clusters (voids, C_{VC}), solute(G) (C_G), solute(G)-interstitial pairs (C_{IG}), solute(M) (C_M), solute(M)-vacancy pairs (C_{VM}), solute-point defect clusters (C_{MC}), the total interstitials in interstitial type dislocation loops (R_{IC}), the total vacancies in voids (R_{VC}), the total point defects in solute-point defect clusters (R_{MCP}), and the total solutes in solute-vacancy clusters (R_{MCM}). For defect clusters, the average size is considered. The rate equations are expressed as follows:

$$\frac{dC_I}{dt} = P_I - 2Z_{I\_I}M_IC_I^2 - Z_{I\_V}(M_I+M_V)C_IC_V - Z_{I\_G}M_IC_IC_G - Z_{I\_2I}M_IC_{2I}C_I + Z_{2I\_V}M_{2I}C_{2I}C_V + 2Z_{BI\_2I}M_IB_IC_{2I} + Z_{2I\_VM}M_{2I}C_{VM}C_{2I} + Z_{2I\_G}M_{2I}C_GC_{2I} - Z_{I\_2V}M_IC_{2V}C_I + Z_{BI\_IG}M_IB_{IG}C_{IG} - Z_{I\_IG}M_IC_{IG}C_I - Z_{I\_VM}M_IC_{VM}C_I - Z_{I\_SI}M_IS_IC_I - Z_{I\_SV}M_IS_VC_I - Z_{I\_SMC}M_IS_{MC}C_I - M_IC_IC_S - N_IP_{IC},$$

$$\frac{dC_V}{dt} = P_V - Z_{I\_V}(M_I+M_V)C_IC_V + Z_{I\_2V}M_IC_{2V}C_I - 2Z_{V\_V}M_VC_V^2 - Z_{V\_M}M_VC_VC_M - Z_{2I\_V}M_{2I}C_VC_{2I} - Z_{V\_2V}M_VC_VC_{2V} + 2Z_{BV\_2V}M_VB_VC_{2V} - Z_{2V\_M}M_{2V}C_MC_{2V} - Z_{IG\_V}M_VC_VC_{IG} - Z_{IG\_2V}M_{IG}C_{2V}C_{IG} - Z_{V\_VM}M_{VM}C_{VM}C_V - Z_{BV\_VM}B_{VM}M_VC_{VM} - Z_{V\_SI}M_VS_IC_V - Z_{V\_SV}M_VS_VC_V - Z_{V\_SMC}M_VC_VS_{MC} - M_VC_VC_S - N_VP_{VC},$$

$$\frac{dC_M}{dt} = Z_{I\_VM}M_IC_{VM}C_I - Z_{V\_M}M_VC_MC_V + Z_{2I\_VM}M_{2I}C_{VM}C_{2I} - Z_{2V\_M}M_{2V}C_MC_{2V} + Z_{VM\_2V}M_{2V}C_{2V}C_{VM} + Z_{V\_VM}M_{VM}C_{VM}C_V + Z_{IG\_VM}M_{IG}C_{VM}C_{IG} + Z_{BV\_VM}B_{VM}M_VC_{VM} + 2Z_{VM\_VM}M_{VM}C_{VM}^2 + Z_{VM\_SV}M_{VM}S_VC_{VM} + Z_{VM\_SMV}M_{VM}S_{MC}C_{VM},$$

$$\frac{dC_G}{dt} = -Z_{I\_G}M_IC_GC_I - Z_{2I\_G}M_{2I}C_GC_{2I} + Z_{I\_IG}M_IC_{IG}C_I + Z_{IG\_2V}M_{IG}C_{2V}C_{IG} + Z_{IG\_VM}M_{IG}C_{VM}C_{IG} + Z_{BI\_IG}B_{IG}M_IC_{IG} + Z_{IG\_V}M_VC_VC_{IG} + Z_{IG\_SV}M_{IG}S_VC_{IG} + Z_{IG\_SMC}M_{IG},$$

$$\frac{dC_{2I}}{dt} = Z_{I\_I}M_IC_I^2 - Z_{I,2I}M_IC_{2I}C_I + Z_{I,IG}M_IC_{IG}C_I - Z_{2I,V}M_{2I}C_VC_{2I} + Z_{2I\_G}M_{2I}C_{2I}C_G + 2Z_{2I\_2I}M_{2I}C_{2I}C_{2I} - Z_{BI\_2I}M_IB_GC_{2I} - Z_{2I\_2V}M_{2I}C_{2V}C_{2I} - Z_{2I\_IG}M_{2I}C_{IG}C_{2I} - Z_{2I\_VM}M_{2I}C_{VM}C_{2I} - Z_{2I\_SI}M_{2I}S_IC_{2I} - Z_{2I\_SV}M_{2I}S_VC_{2I} - Z_{2I\_SMC}M_{2I}S_{BMC}C_{2I} - M_{2I}C_{2I}C_S,$$

$$\frac{dC_{2V}}{dt} = -Z_{I\_2V}M_IC_{2V}C_I + Z_{V\_2V}M_VC_V^2 - 2Z_{V\_2V}M_VC_{2V}C_V - Z_{2V\_M}M_{2V}C_{2V}C_M - Z_{2I\_2V}M_{2I}C_{2V}C_{2I} - 2Z_{2V\_2V}M_{2V}C_{2V}^2 - Z_{BV\_2V}M_VB_VC_{2V} - Z_{IG\_2V}M_{IG}C_{2V}C_{IG} - Z_{VM\_2V}M_{VM}C_{2V}C_{VM} + Z_{V\_VM}M_{VM}C_{VM}C_V + Z_{VM,VM}M_{VM}C_{VM}^2 - Z_{2V\_SI}M_{2V}S_IC_{2V} - Z_{2V,SV}M_{2V}S_VC_{2V} - Z_{2V,SMC}M_{2V}S_{MC}C_{2V} - M_2C_{2V}C_S,$$

$$\frac{dC_{IG}}{dt} = -Z_{I\_IG}M_IC_{IG}C_I - Z_{I\_G}M_IC_GC_I + Z_{IG\_V}M_{IG}C_VC_{IG} - Z_{2I\_IG}M_{2I}C_{IG}C_{2I} + Z_{I\_G}M_{2I}C_GC_{2I} - Z_{IG\_2V}M_{IG}C_{2V}C_{IG} - Z_{BI\_IG}B_{IG}M_IC_{IG} - Z_{IG\_VV}M_{IG}C_{VM}C_{IG} - Z_{IG,SI}M_{IG}S_IC_{IG} - Z_{IG\_SV}M_{IG}S_VC_{IG} - Z_{IG,SMC}M_{IG}S_{MC}C_{IG} - M_{IG}C_{IG}C_S,$$

$$\frac{dC_{VM}}{dt} = -Z_{I\_VM}M_IC_{VM}C_I + Z_{V\_M}M_VC_MC_V - Z_{V\_VM}M_VC_{VM}C_V - Z_{2I\_VM}M_{2I}C_{VM}C_{2I} - Z_{VM\_2V}M_{2V}C_{2V}C_{VM} + Z_{2VI\_M}M_{2V}C_MC_{2V} - Z_{IG\_VM}M_{IG}C_{VM}C_{IG} - 2Z_{VM\_VM}M_{VM}C_{VM}^2 - Zb_{V\_VM}M_VC_{VM} - Z_{VM\_SI}M_{VM}S_IC_{VM} - Z_{VM\_SV}M_{VM}S_VC_{VM} - Z_{VM\_SMC}M_{VM}S_{MC}C_{VM} - M_{VM}C_{VM}C_S,$$

$$\frac{dC_{IC}}{dt} = P_{IC} + Z_{I\_2I}M_IC_{2I}C_I + Z_{2I\_2I}M_{2I}C_{2I}^2 + Z_{2I\_IG}M_{2I}C_{IG}C_{2I} - Z_{IG\_SI}M_{IG}S_IC_{IG} - Z_{VM\_SI}M_{VC}S_IC_{VM},$$

$$\frac{dC_{VC}}{dt} = P_{VC} + Z_{V\_2V}M_VC_{2V}C_C + Z_{2V\_2V}M_{2V}C_{2V}^2 + Z_{VM\_2V}M_{VM}C_{2V}C_{VM},$$

$$\frac{dC_{MC}}{dt} = Z_{2I\_IG}M_{2I}C_{IG}C_{2I} + Z_{IG\_SI}M_{IG}S_IC_{IG} + Z_{VM\_SI}M_{VM}S_IC_{VM},$$

$$\frac{dR_{IC}}{dt} = N_IP_{IC} + Z_{I\_SI}M_IS_IC_I + 3Z_{I\_2I}M_IC_{2I}C_I - Z_{V\_SI}M_VS_IC_V + 4Z_{2I\_2I}M_{2I}C_{2I}^2 + 2Z_{2I\_SI}M_{2I}S_IC_{2I} + 4Z_{2I\_IG}M_{2I}C_{IG}C_{2I} - 2Z_{2V\_SI}M_{2V}S_IC_{2V} - (R_{IC}/C_{IC}Z_{IG\_SI})M_{IG}S_IC_{IG} + 2Z_{2IG\_SI}M_{IG}S_IC_I - (R_{IC}/C_{IC})Z_{VM\_SI}M_{VM}S_IC_{VM},$$

$$\frac{dR_{VC}}{dt} = N_VP_{VC} - Z_{I\_SV}M_IS_VC_I + 3Z_{V\_2V}M_VC_{2V}C_V + Z_{V\_SV}M_VS_VC_V - 2Z_{2I\_SV}M_{2I}S_VC_{2I} + 4Z_{2V\_2V}M_{2V}C_{2V}^2 + 2Z_{2V\_SV}M_{2V}S_VC_{2V} + 3Z_{VM\_2V}M_{VM}C_{2V}C_{VM} - Z_{IG\_SV}M_{IG}S_VC_{IG} + Z_{VM\_SV}M_{VM}S_VC_{VM},$$

$$\frac{dR_{MCP}}{dt} = Z_{I\_SMC}M_IS_{MC}C_I - Z_{V\_SMC}M_VS_{MC}C_V + 2Z_{2I\_SMC}M_{2I}S_{MC}C_{2I} + 3Z_{2I\_IG}M_{2I}C_{IG}C_{2I} - 2Z_{2V\_SMC}M_{2V}S_{MC}C_{2V} + (R_{IC}/C_{IC} + 1)Z_{IG\_SI}M_{IG}S_IC_{IG} + Z_{IG\_SMC}M_{IG}S_{MC}C_{IG} + (R_{IC}/C_{IC} - 1)Z_{VM\_SI}M_{VM}S_IC_{VM} - Z_{VM\_SMC}M_{VM}S_{MC}C_{VM},$$