**Modeling precipitation thermodynamics and kinetics in type 316 austenitic
stainless steels with varying composition as an initial step towards
predicting phase stability during irradiation**

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Abstract

The long-term evolution of precipitates in type 316 austenitic stainless steels at 400 °C has been simulated using a numerical model based on classical nucleation theory and the thermodynamic extremum principle. Particular attention has been paid to the precipitation of radiation-induced phases such as γ' and G phases. In addition to the original compositions, the compositions for radiation-induced segregation at a dose level of 5, 10 or 20 dpa have been used in the simulation. In a 316 austenitic stainless steel, γ' appears as the main precipitate with a small amount of G phase forming at 10 and 20 dpa. On the other hand, G phase becomes relatively dominant over γ' at the same dose levels in a Ti-stabilized 316 austenitic stainless steel, which tends to suppress the formation of γ' . Among the segregated alloying elements, the concentration of Si seems to be the most critical for the formation of radiation-induced phases. An increase in dislocation density as well as increased diffusivity of Mn and Si significantly enhances the precipitation kinetics of the radiation-induced phases within this model.

1. Introduction

Austenitic stainless steels (ASSs) including 304, 316 and 347 grades are widely used for the core components of reactor pressure vessels in light water reactors (LWRs) due to their relatively high strength and good fracture toughness. These components experience significant neutron irradiation fluence during the service lifetime in LWRs, which leads to microstructural changes, phase instability and property degradation of the components [1,2]. In addition, compositional changes due to radiation-induced segregation (RIS) are usually observed in irradiated ASS components [3]. Radiation-induced defects diffuse to sinks such as grain boundaries during irradiation, and the excess defect fluxes to sinks causes non-equilibrium segregation of solute atoms due to an inverse Kirkendall effect and an interstitial association mechanism [4]. The RIS behavior of Cr and Ni in irradiated ASSs is well established, in which Cr depletes at grain boundaries while Ni is enriched [5,6]. The Cr and Ni segregation behavior is well explained by a modified inverse Kirkendall effect [7]. Si is also generally observed to enrich at grain boundaries in irradiated ASSs, which is usually explained by an interstitial association mechanism [8]. In addition to grain boundaries, RIS has been observed at voids and Frank loops in irradiated ASSs [9].

In LWR environments, the precipitation of second phases can take place under the combined effects of temperature, increased defect concentration and locally varying compositional domains due to RIS, and the second phase precipitate distributions clearly influence the hardening and embrittlement of the components [3]. Radiation-induced phases (RIPs) are precipitates that appear only under irradiation but not for corresponding thermal conditions [3]. γ' and G phases are the typical RIPs observed in ASSs [10-20]. Of the various grades of ASSs, 316 may be the most susceptible to the formation of γ' and G phases due to

the higher Ni content. γ is an A_3B type intermetallic compound with an ordered fcc crystal structure ($L1_2$) and usually appears as Ni_3Si in irradiated ASSs. Radiation-induced γ has been observed to form near Frank loops in a temperature range between 300 and 500 °C in both 316 and Ti-stabilized 316 ASSs [17]. G phase is a complex silicide, $M_6Ni_{16}Si_7$ ($M = Ti, Nb, Mn, Fe$ and/or Cr) with an fcc crystal structure ($A1$). Although G phase as a RIP was reported first in irradiated Ti- and Nb-stabilized 316 ASSs [12,17], further studies revealed that G phase is also observed, although at relatively low volume fractions, during irradiation in a 316 ASS [13,14,16,17]. Radiation-induced G phase has been observed to form near voids in a temperature range of 400-650 °C [17]. Further, a recent study indicates Ni and Si segregation to dislocation lines and loops in a proton irradiated 304 ASS, and that the dislocation can act as a RIP nucleation site [21].

In addition to experimental observations, there has been recent effort to simulate the precipitation kinetics in ASSs during aging by combining classical nucleation theory with growth equations from diffusion theory or the thermodynamic extremum principle [22-24]. These efforts were successful in providing a reasonably accurate prediction of the precipitation sequence, the precipitate phase fraction and precipitate size in ASSs, compared with experimental observations. However, no currently published study has predicted the precipitation kinetics of RIPs in ASSs.

The purpose of this study is to simulate the long-term evolution of precipitates in 316 and Ti-stabilized 316 ASSs considering the RIS effect using a numerical model based on classical nucleation theory and evolution equations derived from the thermodynamic extremum principle. Special attention is given to the precipitation kinetics of γ and G phases.

2. Computational details

The composition of a 316 ASS for the simulation was determined according to Refs. [13,16], and is shown in Table 1. A Ti-stabilized 316 ASS contains a small amount of Ti to prevent the formation of $M_{23}C_6$, which is responsible for sensitization. The composition of solute elements as a result of RIS at 5, 10 or 20 dpa was determined according to the measurements of grain boundary compositions in irradiated 316 ASSs in the literature [20,25-45]. The content of Cr, Ni and Si was taken from the average values of the data at various dose levels compiled in the literature [25-45]. It should be noted that the Si content measured by atom probe tomography (APT) [37-39,42,44] is usually larger than that measured by transmission electron microscopy (TEM) combined with energy dispersive X-ray spectroscopy (EDS) [25,26,32]. This is presumably because APT provides a higher spatial resolution than TEM-EDS, which can detect more localized segregation of Si. The content of Mn, Mo and Ti was set constant irrespective of radiation dose using the results of Jiao and Was [20], Ohnuki et al. [25], Kenik et al. [27], Kato et al. [29], Kasahara et al. [32], Fukuya [42] and Hatakeyama et al. [44], as the dependence of the segregation behavior of Mn, Mo and Ti on dose has not been well established.

In addition to the calculation of the phase equilibria (e.g., equilibrium precipitation fractions) in type 316 ASSs, the simulation of the precipitate evolution in these steels has been performed with the thermo-kinetic software package MatCalc (version 5.61, release 0.006) developed by Kozeschnik et al. [46,47]. This model treats the kinetics of microstructural processes based on classical nucleation theory and evolution equations for the radius and composition of each precipitate derived from the thermodynamic extremum principle. The details of the basic principles composing MatCalc are given in Ref. [48]. We

should mention that the thermodynamic calculation and the kinetic simulation are not spatially resolved, and thus are not performed for all regions in the steel. Instead, we have selected representative compositions that represent localized spatial regions, with different compositions than the bulk as a result of RIS. In such regions near strong defect sinks such as grain boundaries, dislocations or voids, RIS can provide a thermodynamic driving force for the formation of RIPS as locally stable phases. During the calculation and simulation, the thermodynamic and kinetic data for all phases, except for γ' and G phases, were calculated from the MatCalc thermodynamic database 'mc_fe' (version 2.003) and the MatCalc mobility database 'mc_fe' (version 2.006), respectively [49]. The thermodynamic data for γ' was taken from the thermodynamic assessment of the Ni-Si-Ti system by Tokunaga et al. [50]. G phase was modeled as $(\text{Mn,Ti})_6\text{Ni}_{16}\text{Si}_7$ considering the solubility between Mn and Ti. The thermodynamic data for $\text{Mn}_6\text{Ni}_{16}\text{Si}_7$ and $\text{Ti}_6\text{Ni}_{16}\text{Si}_7$ were taken from the assessments of Hu et al. [51] and Tokunaga et al. [50], respectively. The thermodynamic parameters for γ' and G phases used in the present work are listed in Table 2.

The matrix phase defined in the simulation is austenite. The partial transformation of the matrix into ferrite at low temperatures, which is known to be accelerated under irradiation conditions [17], was not considered in the model. Also, it is assumed that the composition is maintained uniform across the matrix and does not change with time. The grain size of austenite was assumed to be 50 μm , which is in the range obtained after conventional thermo-mechanical treatments of a 316 ASS [52]. The dislocation density of the austenite matrix is assumed to be 10^{13} m^{-2} , according to the measured data on solution annealed ASSs [53].

M_{23}C_6 , M_6C , MX, Laves, χ , σ , γ' and G phases were included in the simulation as possible precipitates. For the crystal structure and chemical composition of these precipitates, see the review by Sourmail [54]. The nucleation sites of the χ and σ phases were set to be

grain boundaries, those of Laves phase and MX were set to dislocations and those of $M_{23}C_6$ and M_6C were set to both grain boundaries and dislocations, according to information in the literature [22,55]. The nucleation sites of γ' and G phases were set to dislocations, as γ' and G phases are known to nucleate mainly at Frank loops and voids, respectively, inside grains [17]. Although a recent study [20] reported that there is a tendency for the formation of Ni/Si-rich clusters before the formation of γ' and G phases, this clustering tendency was not reflected in this simulation due to the limit of classical nucleation theory. The diffusivity ratio between precipitate and matrix, which influences the kinetics of the precipitate composition evolution, was set to 1.0 for all precipitates.

The interfacial energy of the precipitates is an important factor in determining their nucleation and growth rate. In MatCalc, the interfacial energy is calculated from thermodynamic data, based on the generalized broken bond model [56]. In this study, the calculated interfacial energy values of the precipitates were used without adjustment. Instead, the nucleation constants of $M_{23}C_6$, χ , σ and Laves phases were adjusted to 10^{-4} , 10^{-14} , 10^{-10} and 10^{-1} , respectively, to reproduce the known precipitation kinetics in a 316 ASS at 600 °C reported by the National Institute for Materials Science [57].

The heat treatment started with a solution treatment at 1250 °C. It was initially assumed that all elements were homogeneously distributed in the matrix and no precipitates existed. The simulations of the precipitation kinetics of the ASS with modified compositions corresponding to RIS observations, were also assumed to follow the same thermal history, including the 1250°C solution treatment. After the solution treatment, the steels were cooled linearly down to room temperature in 30 min and then subsequently heated up to 400 °C in 30 min. The aging was simulated at 400 °C for 100,000 h. Most of the precipitates, except MX, form during aging, due to the long time required for nucleation.

3. Results and discussion

Fig. 1 shows the calculated equilibrium mole fraction of second phase precipitates in a 316 ASS, assuming uniform compositions associated with RIS at 0, 5, 10 or 20 dpa as a function of temperature, together with that of austenite and ferrite. For the unirradiated 316 ASS (0 dpa), the major precipitates are Laves and σ phases, together with a small amount of $M_{23}C_6$, which is stable throughout the temperature range. A small fraction of γ' starts to form below 360 °C. Although the austenite-ferrite (γ - α) transformation starts at about 550 °C, a significant fraction of austenite still remains at 300°C (Fig. 1a). At a modified composition associated with RIS at 5 dpa, the fraction of γ' remarkably increases, while the fraction of Laves and σ phases decreases (Fig. 1b). The austenite-ferrite transformation temperature decreases by about 50 °C due to the enrichment of Ni, which is an austenite stabilizer. As the dose level increases further, the fraction of γ' continues to increase and G phase appears due to the enrichment of Ni and Si (Figs. 1c and d). On the other hand, the fraction of σ phase decreases slightly. The austenite-ferrite transformation temperature increases due to the enrichment of Si, which is a strong ferrite stabilizer. $M_{23}C_6$ tends to remain unaffected in spite of RIS. As shown in Fig. 2, the calculated equilibrium phase fraction in a Ti-stabilized 316 ASS is not significantly different from that in a 316 ASS, except that MX, which is a titanium carbonitride containing a small amount of Cr, partially replaces $M_{23}C_6$.

The simulated precipitate evolution kinetics in a 316 ASS, assuming the uniform compositions associated with RIS at 0, 5, 10 or 20 dpa as a function of aging time at 400 °C is shown in Fig. 3. For the unirradiated 316 ASS composition (0 dpa), only a small fraction of

$M_{23}C_6$ forms at about 10,000 h of aging (Fig. 3a). The precipitation kinetics of $M_{23}C_6$ is slightly reduced at the RIS composition at 5 dpa due to the depletion of Cr, with no indication of the formation of other phases (Fig. 3b). At the RIS composition at 10 dpa, a small amount of γ' starts to form after 10,000 h of aging (Fig. 3c) together with $M_{23}C_6$. Also, a trace amount of G phase starts to appear after 10,000 h of aging. Comparing the results between 5 and 10 dpa, the RIPs (γ' and G phases) can kinetically appear at the RIS composition around 10 dpa, for which the Cr concentration has been reduced below 13 wt% and the Ni and Si concentrations have increased above 20 and 4 wt%, respectively. This is consistent with experimental findings in the literature [3,13,16,17], reporting that γ' and G phases start to form in irradiated 316 ASSs before 10 dpa. The main difference in the assumed RIS composition between 5 and 10 dpa is the Si content, which increases from 3 to 5 wt% at 5 and 10 dpa, respectively. The Si content seems to be a critical variable in the formation of γ' and G phases, as the small difference in Si content significantly influences the precipitation kinetics of these phases. At the RIS composition at 20 dpa, the precipitation kinetics of γ' is accelerated with a start time at 2,000 h of aging (Fig. 3d). In addition, the fraction of G phase significantly increases next to that of γ' . The prediction that γ' appears as a dominant RIP over G phase in irradiated 316 ASS is consistent with experimental observations [10-17].

Fig. 4 shows the simulated precipitate evolution kinetics in a Ti-stabilized 316 ASS with the RIS compositions at 10 or 20 dpa. At the RIS composition at 10 dpa, MX rapidly forms during the cool to room temperature from the solution treatment at 1250 °C (Fig. 4a). G phase appears as the main second-phase precipitate, which starts to form after 3,000 h of aging and tends to saturate after 60,000 h of aging. On the other hand, the formation of γ' is significantly suppressed compared with a 316 ASS (Fig. 3b). At the assumed RIS composition at 20 dpa, the fraction of γ' significantly increases, while both G and γ' phases exhibit a similar

precipitation behavior (Fig. 4b). On the whole, it is predicted that the addition of Ti to a 316 ASS induces enhanced formation of G phase, while suppressing γ' formation, which is in agreement with experimental observations [12,13,17].

The dose rate for type 316 ASSs in experimental reactors such as HFIR and EBR-II is approximately 10^{-6} dpa/s [3]. Therefore, it is estimated to take about 2,800 h for the radiation dose to reach 10 dpa in type 316 ASSs in these environmental conditions, at which point the RIPs are observed. In addition, the specimens stay below 5 dpa for the initial 1,400 h, which might not make a big contribution to the formation of RIPs. However, the present simulation results indicate that it takes more than 10,000 h for the RIPs to form at significant fraction (Figs. 3 and 4). Therefore, we need to consider factors that can accelerate the formation of RIPs in the simulations. It has been known that the matrix dislocation density in type 316 ASSs increases during irradiation. Zinkle et al. [53] presented the variation of the dislocation density in type 316 ASSs with dose in the temperature range between 385 and 400 °C by compiling the experimental data in the literature. The dislocation density drastically increases with dose below 5 dpa and exhibits a saturation tendency for doses greater than 15 dpa. The dislocation density can be estimated to be 0.8×10^{15} , 1.5×10^{15} and 2×10^{15} m⁻² at 5, 10 and 20 dpa, respectively. The increase in dislocation density is expected to provide more nucleation sites and increased density of fast diffusion paths for precipitation [58]. The simulation of precipitate evolution has thus been performed using these dislocation densities in irradiated 316 ASSs. Details of the implementation of the pipe diffusion effect into MatCalc simulations are reported in Ref. [58]. Fig. 5 shows the simulated precipitate evolution kinetics in 316 and Ti-stabilized 316 ASSs with the RIS compositions at 10 dpa after incorporating the increased dislocation density (1.5×10^{15} m⁻²). In the 316 ASS, the precipitation kinetics of γ' , G phase and M₂₃C₆ is drastically accelerated (Fig. 5a), compared with the results of a dislocation

density of $1 \times 10^{13} \text{ m}^{-2}$ (Fig. 3c). γ' starts to form after 20 h of aging and is fully precipitated after 1,000 h of aging. Although the precipitation kinetics of both the γ' and G phases are also significantly enhanced in the Ti-stabilized 316 ASS (Fig. 5b), γ' is more influenced by the increase in dislocation density. The fraction of γ' exceeds that of G phase after 700 h of aging, which is in contrast to the result at a dislocation density of $1 \times 10^{13} \text{ m}^{-2}$, which predicts that G phase is relatively dominant over γ' (Fig. 4a). The increase in dislocation density appears to accelerate the rate at which the thermodynamic equilibrium is reached by providing fast diffusion paths of alloying elements, predicting that γ' is dominant over G phase even in the Ti-stabilized 316 ASS (Fig. 2b).

The simulated average precipitate size in 316 and Ti-stabilized 316 ASSs with the RIS compositions at 10 dpa and the increased dislocation density ($1.5 \times 10^{15} \text{ m}^{-2}$) is shown in Fig. 6. In the 316 ASS, the average size of M_{23}C_6 increases relatively rapidly with increasing aging time, reaching about 80 nm. γ' and G phases are predicted to grow in size up to about 50 and 80 nm, respectively (Fig. 6a). This size is consistent with the range reported in the literature [16,17], which is 5-50 nm and 10-250 nm for γ' and G phases, respectively. In the Ti-stabilized ASS, the size of MX remains almost constant (about 30 nm) during aging (Fig. 6b). The size of G phase is maintained at about 10 nm after 20 h of aging and gradually increases after 10,000 h of aging, while the size of γ' relatively rapidly increases after 30 h of aging, reaching about 50 nm at 100,000 h of aging. The simulated compositional change of G phase in the Ti-stabilized 316 ASS with the RIS composition at 10 dpa is presented in Fig. 7. After 300 h of aging, the Mn content in G phase tends to increase with a big fluctuation around 2,000 h of aging, while the composition is maintained to be Ti-rich at the initial stage. This fluctuation seems to be directly related with the formation of γ' . While the content of Si in the matrix decreases as γ' continues to form, it does not change after the full precipitation of γ' .

During this transient state where the content of Si changes in the matrix, G phase seems to adapt its chemical composition to the Mn-rich direction to respond to the change in chemical environment. The composition returns to the original trend after the transient state. The overall trend where the Mn content in G phase steadily increases during the whole aging period seems to be associated with the approach to thermodynamic equilibrium, which predicts that the equilibrium Mn and Ti contents in G phase at the 10 dpa RIS composition at 400 °C are about 21 and 5×10^{-3} at%, respectively.

In addition to changes in dislocation density under irradiation, diffusivity could be a factor that accelerates the precipitation of γ' and G phases. The bulk diffusivity of alloying elements has been found to significantly increase during irradiation due to the formation of radiation defects such as vacancies and interstitial atoms, which is known as radiation-enhanced diffusion [3]. Akasaka et al. [59] measured the bulk diffusivity of Cr and Ni in a 316 ASS at 370, 460 and 540 °C using miniature diffusion couples irradiated in an experimental reactor. They found that the bulk diffusivity of Cr and Ni at 370 °C increases 10^6 times compared with that in an unirradiated 316 ASS. Unfortunately, the diffusivity of other alloying elements in irradiated austenitic steels has not yet been reported. The simulation of precipitate evolution kinetics with increasing the Cr and Ni diffusivity by a factor of 10^6 is shown in Fig. 8a, and indicates that the precipitation kinetics of γ' and $M_{23}C_6$ is slightly enhanced. However, if the diffusivity of Mn and Si is also increased by the same order of magnitude, then the precipitation kinetics of γ' and G phases is drastically accelerated as shown in Fig. 8b. As soon as the aging begins with these simulated conditions, both γ' and G phases start to form and exhibit a saturation behavior before 1,000 h of aging. Therefore, we conclude that the diffusivity of Mn and Si, with significantly smaller concentrations relative to Ni, appear to play a rate controlling role in determining the precipitation kinetics

of the RIPs, especially for the G and γ' phases. Therefore, improved assessments of the radiation enhanced diffusion of Mn and Si represent an important need for modeling RIP in ASS.

4. Conclusions

Numerical simulation of the long-term precipitate evolution in type 316 austenitic stainless steels (ASSs) at 400 °C has been performed with special attention to γ' and G phases that are typical radiation-induced phases (RIPs), using evolution equations based on a combination of classical nucleation theory and the thermodynamic extremum principle. To reflect the irradiation effect, the measured radiation-induced segregation (RIS) compositions at grain boundaries irradiated to 5, 10 or 20 dpa reported in the literature, as well as the original compositions, have been used in the simulation. For compositions at radiation doses greater than 5 dpa, γ' and G phases are predicted to be thermodynamically stable at 400 °C due to the enrichment of Ni and Si occurring by RIS. In the simulation of the precipitation kinetics, γ' and G phases form together with a small amount of $M_{23}C_6$ at 10 and 20 dpa, although only $M_{23}C_6$ is predicted to appear at 0 and 5 dpa. The content of Si appears to play a crucial role in the formation of γ' and G phases. Although γ' is dominant over G phase in the 316 ASS, G phase appears as the major RIP in a Ti-stabilized ASS. It is found that an increase in dislocation density, as is observed to occur in irradiated 316 ASSs, significantly enhances the precipitation kinetics of γ' and G phases, as well as $M_{23}C_6$ and M_6C . Further, increases in the diffusivity of Mn and Si, as expected due to radiation enhanced diffusion, drastically accelerates the precipitation kinetics of both phases, even more so than increases in the Cr or

Ni diffusivities. The predicted average precipitate size of the RIPs is in reasonable agreement with experimental observations in the literature.

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Appendix

In the precipitation kinetics approach implemented in MatCalc, the evolution of the thermodynamic system is simulated in the framework of the Kampmann-Wagner model [60]. Accordingly, the total time history is broken into adequately small, isothermal segments [47]. Precipitates of equal size and chemical composition are grouped in classes, for each of which the evolution in radius and composition is calculated according to the rate equations derived from the thermodynamic extremum principle [46]. In this approach, the nucleation kinetics of precipitates was calculated from classical nucleation theory extended for multicomponent systems [48]. Accordingly, the transient nucleation rate J is given as:

$$J = N_0 Z \beta^* \cdot \exp\left(\frac{-G^*}{kT}\right) \cdot \exp\left(\frac{-\tau}{t}\right) \quad (1)$$

J describes the rate at which nuclei are created per unit volume and time. N_0 represents the total number of potential nucleation sites. The Zeldovich factor Z takes into account that the nucleus is destabilized by thermal excitation as compared to the inactivated state, and is given:

$$Z = \left[\frac{-1}{2\pi kT} \frac{\partial^2 \Delta G}{\partial n^2} \Big|_{n^*} \right]^{\frac{1}{2}}. \quad (2)$$

ΔG represents the total free energy change on nucleus formation, n is the number of atoms in the nucleus, k is the Boltzmann constant and T is temperature. The derivative of ΔG is taken at the critical nucleus size n^* .

The atomic attachment rate β^* takes into account the long-range diffusive transport of atoms, which is needed for nucleus formation if the chemical composition of the matrix is different from the chemical composition of the precipitate. A suitable multicomponent equation for β^* has been derived by Svoboda et al. [48], and is given:

$$\beta^* = \frac{4\pi \rho^{*2}}{a^4 \Omega} \left[\sum_{i=1}^n \frac{(c_{ki} - c_{0i})^2}{c_{0i} D_{0i}} \right]^{-1}, \quad (3)$$

where ρ^* is the critical nucleation radius, a is the atomic distance, Ω is the molar volume, c_{ki} and c_{0i} are the element concentrations in precipitate with index k and matrix, respectively, and D_{0i} is the diffusivity of element i in the matrix. The incubation time τ is given:

$$\tau = \frac{1}{2\beta^* Z^2} , \quad (4)$$

and the critical energy for nucleus formation G^* is:

$$G^* = \frac{16\pi}{3} \frac{\gamma^3}{(\Delta G_{\text{vol}})^2} , \quad (5)$$

with the interfacial energy γ and the volume free energy change ΔG_{vol} .

It is important to note that G^* is an essential quantity in nucleation theory, when compared to the other quantities occurring in Eq. (1). G^* contains the cube of the interfacial energy over the square of the volume free energy change ΔG_{vol} . Since G^* appears in the exponent of the nucleation rate Eq. (1), small changes in γ and/or ΔG_{vol} can lead to huge variations in J , particularly if G^* is of the order of kT . In the present case, these quantities change steadily during continuous cooling and in the course of the precipitation reaction, since both are functions of temperature and chemical composition of matrix and precipitate. These interactions are discussed in detail in the following sections.

Once a precipitate is nucleated, its further growth is evaluated based on the evolution equations for the radius and composition of the precipitate derived by Svoboda et al. [48] based on a mean-field approach utilizing the thermodynamic extremal principle. Accordingly, the growth rate $\dot{\rho}_k$ and the rate of change of chemical composition $\dot{c}_{ki}$ of the precipitate with index k are obtained from solution of the linear system of equations:

$$A_{ij}y_j = B_i, \quad (6)$$

where the variable y_j represents the rates $\dot{\rho}_k$ and $\dot{c}_{ki}$, as well as the Lagrange multipliers from the stoichiometry constraints (see Ref. [48]). The index i comprises the variables for radius and chemical composition of the precipitate as well as the stoichiometry boundary conditions implied from the crystal structure of the precipitate phase. The system of Eq. (6) is solved for each precipitate k separately. The full expressions for the coefficients A_{ij} and B_i , as used in the present work, are given in Ref. [47]. The numerical time integration of $\dot{\rho}_k$ and $\dot{c}_{ki}$ is performed in the software MatCalc, based on the classical numerical Kampmann–Wagner approach [60], and is described in detail Ref. [46].

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Table 1. Compositions of type 316 austenitic stainless steels (wt%).

	Fe	Cr	Ni	Mo	Mn	Si	(Ti)	C	N
316	Bal.	17	13	2.2	1.8	0.6	(0.2)	0.05	0.03
Segregation at 5dpa	Bal.	14±2	18±2	1	1	3±2	(0.5)	0.05	0.03
Segregation at 10 dpa	Bal.	12±1	21±4	1	1	5±1.5	(0.5)	0.05	0.03
Segregation at 20 dpa	Bal.	11±2	24±2	1	1	6	(0.5)	0.05	0.03

Table 2. Thermodynamic parameters for γ' and G phases.

Thermodynamic parameters (J/mol)	Ref.
γ': model $(\text{Ni},\text{Si})_{0.75}(\text{Ni},\text{Si})_{0.25}$	
${}^oG_{\text{Ni:Ni}} = {}^oG_{\text{Ni}}^{\text{fcc}}$	
${}^oG_{\text{Ni:Si}} = 0.75 \cdot {}^oG_{\text{Ni}}^{\text{fcc}} + 0.25 \cdot {}^oG_{\text{Si}}^{\text{diamond}} - 49886.12 + 5.53901T$	
${}^oG_{\text{Si:Ni}} = 0.25 \cdot {}^oG_{\text{Ni}}^{\text{fcc}} + 0.75 \cdot {}^oG_{\text{Si}}^{\text{diamond}} + 20600$	[50]
${}^oG_{\text{Si:Si}} = {}^oG_{\text{Si}}^{\text{fcc}}$	
${}^0L_{\text{Ni,Si:Ni}} = {}^0L_{\text{Ni,Si:Si}} = 50000$	
${}^0L_{\text{Ni:Ni,Si}} = {}^0L_{\text{Si:Ni,Si}} = 3016.11 - 6.08310T$	
G phase: model $(\text{Ti},\text{Mn})_6\text{Ni}_{16}\text{Si}_7$	
${}^oG_{\text{Ti:Ni:Si}} = 6 \cdot {}^oG_{\text{Ti}}^{\text{hcp}} + 16 \cdot {}^oG_{\text{Ni}}^{\text{fcc}} + 7 \cdot {}^oG_{\text{Si}}^{\text{diamond}} - 1585300.40$	[50]
$- 130.88101T$	
${}^oG_{\text{Mn:Ni:Si}} = 6 \cdot {}^oG_{\text{Mn}}^{\text{cbcc}} + 16 \cdot {}^oG_{\text{Ni}}^{\text{fcc}} + 7 \cdot {}^oG_{\text{Si}}^{\text{diamond}} - 1397408.413$	[51]

Figure captions

Fig. 1. Calculated equilibrium mole fraction of phases in a 316 ASS with the RIS compositions at (a) 0, (b) 5, (c) 10 and (d) 20 dpa as a function of temperature.

Fig. 2. Calculated equilibrium mole fraction of phases in a Ti-stabilized 316 ASS with the RIS compositions at (a) 0 and (b) 10 dpa as a function of temperature.

Fig. 3. Simulated precipitate evolution kinetics in a 316 ASS with the RIS compositions at (a) 0, (b) 5, (c) 10 and (d) 20 dpa as a function of aging time at 400 °C.

Fig. 4. Simulated precipitate evolution kinetics in a Ti-stabilized 316 ASS with the RIS compositions at (a) 10 and (b) 20 dpa as a function of aging time at 400 °C.

Fig. 5. Simulated precipitate evolution kinetics in (a) 316 and (b) Ti-stabilized 316 ASSs with the RIS compositions at 10 dpa after incorporating the increased dislocation density ($1.5 \times 10^{15} \text{ m}^{-2}$) as a function of aging time at 400 °C.

Fig. 6. Simulated average precipitate size in (a) 316 and (b) Ti-stabilized 316 ASSs with the RIS compositions at 10 dpa after incorporating the increased dislocation density ($1.5 \times 10^{15} \text{ m}^{-2}$) as a function of aging time at 400 °C.

Fig. 7. Simulated composition of G phase in Ti-stabilized 316 ASS with the RIS compositions at 10 dpa after incorporating the increased dislocation density ($1.5 \times 10^{15} \text{ m}^{-2}$) as a function of aging time at 400 °C.

Fig. 8. Simulated precipitate evolution kinetics in a 316 ASS with the RIS compositions at 10 dpa after incorporating the increased diffusivity of (a) Cr and Ni and (b) Cr, Ni, Mn and Si as a function of aging time at 400 °C.

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