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Predicting the Irradiation Swelling of Austenitic and Ferritic/Martensitic Steels, Based on the Coupled Model of Machine Learning and Rate Theory

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Abstract: As nuclear structural materials, austenitic and ferritic/martensitic (F/M) steels will face inevitable irradiation swelling when fulfilling a role in nuclear reactors, especially under high-dose irradiation. For this work, a coupled machine learning rate theory (ML-RT) model for the swelling of austenitic and F/M steels was developed. In this model, ML was introduced to predict the steady-state irradiation swelling onset dose (D_{onset}), while the improved RT was developed to simulate the swelling behavior after the incubation period. More than 200 series of data on the D_{onset} of different structures of steel were collected for the ML prediction. The coefficient of determination (R) of the results in ML is more than 0.9. In the RT, the evolutions of the dislocation loop and void were described and calculated rather than using the fitting parameters. Cascade efficiency was employed to describe the cascade process. The coupled ML-RT model was verified with the swelling data from neutron irradiation experiments for various steels. The theoretical results of the swelling peak temperatures and swelling behavior are more accurate and reasonable, compared with those from the previous RT model. Using the ML-RT model, the swelling performance of CLAM steel under neutron irradiation of up to 180 dpa was predicted. The differences between the swelling performance of austenitic steels and F/M steels were analyzed and the differences were mainly associated with the bias. These results will be helpful for evaluating the neutron irradiation swelling behavior of candidate structural materials.

Keywords: austenitic and ferritic/martensitic steels; irradiation swelling; rate theory; machine learning



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1. Introduction

Radiation damage, in the form of void formation in materials, leads to swelling [1] and presents a major challenge to the integrity of structural materials in nuclear reactors. When the swelling volume of the material reaches 5% [2], the material will not be able to maintain its basic performance, resulting in a series of problems in terms of strength, stability, etc., meaning that it cannot meet the needs of engineering design. The structural materials in fusion reactors need to withstand irradiation damage from an accumulated irradiation dose of up to hundreds of dpa. As a potential fusion structural material, reduced-activation ferritic/martensitic (RAFM) steel has relatively good irradiation resistance, but the degree of swelling may still become unacceptable after receiving high-dose neutron radiation in the fusion reactors. Neutron irradiation experiments are expensive and take a tremendous amount of time. Self-ion bombardment experiments are usually used as a surrogate for neutron irradiation. However, there are differences between ion and neutron irradiation, especially in terms of the spatial and temporal distribution of irradiation damage, which results in changes in void radius, density, and depth distribution. In addition, the void formation process is difficult to observe in the experiments. A theoretical model is needed to study the swelling behavior of nuclear structural materials. Specifically, the rate theory

(RT) model is well-suited for long-term swelling behavior monitoring under high-dose irradiation because of its fast calculation speed and long calculable time span.

Regarding the development of the RT model, Brailsford and Bullough et al. proposed a set of formulas in the early 1970s for calculating swelling using the reaction rate [3]. Since then, further research has been carried out addressing different aspects of the phenomenon. Factors controlling the growth of voids and bubbles in irradiated materials have been explored using the classic RT model [4]. In particular, the research has been focused on dislocation loops, network dislocations, and other defect sinks that can noticeably influence void evolution [5]. However, the classic RT model cannot describe the incubation period well. In our model, the incubation period was ignored by setting an initial radius for the void and dislocation loop. Although the improved cluster dynamics method tries to describe the nuclear process, this method is still time-consuming due to the complexity of the realistic scenarios, such as alloying elements, the production of gaseous transmutation products, irradiation segregation, etc. [6]. In addition, the fitting parameters usually used for calculating decrease the credibility of simulated results [7]. In this work, we introduce machine learning (ML) to predict the steady-state swelling onset dose (D_{onset}). As a computational model based on a dataset, ML has already proved its applicability for predicting the D_{onset} in the work of Jin et al. [8]. Based on the numerous experimental results regarding the irradiation swelling of structural materials, ML could calculate and predict the D_{onset} quickly without a physical model. However, due to the lack of experimental swelling data under high-dose conditions, ML cannot provide reasonable predictions of swelling behavior after hundreds of doses.

The classic RT model also meets some problems when simulating neutron irradiation swelling at high temperatures ($>525\text{ }^{\circ}\text{C}$), due to the treatment of the cascade effects by considering the formation of vacancy loops [9]. In fact, the clusters formed during the cascade process do not only contain vacancy loops [10]; thus, the model considering the generation of vacancy loops, to deal with cascade effects, needs to be improved. The interstitial loops play a more important role in swelling at the void evolution stage than vacancy loops [11]. Okita et al. used cascade efficiency to describe the generation of point defects during the cascade process [12]. However, the improved model can only calculate the swelling rate under a certain dose, based on the existing experimental data for voids, so the prediction function of the model is missing. On the other hand, the swelling of austenitic steels has been studied more thoroughly than that of F/M steels. It is necessary to develop the RT model to predict the swelling behaviors of F/M steels.

In this work, a coupled machine learning-rate theory (ML-RT) model for the irradiation swelling of both austenitic and F/M steels was developed based on the ML and classic RT. In this ML-RT model, ML was introduced to predict the D_{onset} , while the improved RT model was used to simulate the swelling behavior demonstrated after the incubation period. Cascade efficiency is employed to describe the production of defects during the cascade process [13], and the evolution of dislocation loops and voids according to dose was considered as well. Using this ML-RT model, the swelling of F/M steels under neutron irradiation of up to hundreds of dpa could be calculated rapidly. The dominant affecting factors for the swelling resistance properties of different materials were analyzed. These results are helpful for evaluating the high-dose neutron irradiation swelling performance of candidate structural materials.

2. Materials and Methods

The coupled machine learning-rate theory (ML-RT) model contains two parts. Calculations for the D_{onset} are mainly based on the ML method and for swelling after the incubation period, they are mainly based on the RT model.

2.1. Machine Learning of Steady-State Swelling Onset Dose

The ML method was used to predict the D_{onset} of structure materials under irradiation. D_{onset} was defined as the intersection point between the steady-state swelling extension

line and the horizontal axis of dose, calculated according to the work of Jin et al. [8]. In this work, the original data is from irradiation experiments in literature, as numerous irradiation experiments have been carried out during the past decades. The material compositions, irradiation dose rate, temperature, dislocation density, and cascade efficiency were used as the features in ML. The compositions include Fe, Cr, Ni, Mn, Mo, Si, W, V, Ta, Ti, etc. The cascade efficiency was introduced to identify the irradiation types (electron, ion, or neutron irradiation). The complete dataset contains more than 200 samples, including those for martensitic steels and F/M steels. Table 1 shows the range of values for each feature in this dataset. If readers are interested in the data, please contact the corresponding author for the complete dataset. The ensemble methods applied in this ML study include random forest regression (RFR), support vector regression (SVR), decision tree regression (DTR), gradient boosting regression (GBR), and k-nearest neighbor regression (KNN). These machine learning algorithms were applied using the scikit-learn 0.22.2 Python package. The prediction reliability of each ensemble method was assessed using the correlation coefficient (R) and the root mean square error (RMSE). The dataset was normalized, then all the data were divided into two groups, the training set (80%) and the testing set (20%).

Table 1. The range of numerical variables in the dataset.

Variables	Min	Max	Variables	Min	Max
Fe/(wt %)	8.0	97.0	V/(wt %)	0	2
Cr/(wt %)	3	24.7	W/(wt %)	0	2.1
Ni/(wt %)	0.0	74.9	Temperature/(K)	500	1013
Si/(wt %)	0.0	1.3	Dose rate/(dpa/s)	8×10^{-9}	0.06
Mn/(wt %)	0.0	15	Dislocation density/(m^{-2})	5×10^{13}	8.5×10^{15}
Mo/(wt %)	0.0	2.8	Cascade efficiency	0.01	0.3
Ta/(wt %)	0.0	0.36	Dose(dpa)	0.2	120

In the classic RT model, due to the setting of the initial radiuses of the voids and dislocation loops, the calculation starts from an initial dose that can be denoted as $D_{i,RT}$. If the D_{onset} is obtained by ML, then the $D_{i,RT}$ can be calculated by:

$$D_{i,RT} = D_{onset} + 0.1\% / R_s \quad (1)$$

where 0.1% is the approximate swelling value that is calculated by the following formula:

$$S_w = \frac{4}{3} \pi r_v^3 C_v \quad (2)$$

where S_w is the material swelling value, C_v is the concentration of voids under irradiation and can be acquired from experiments using Transmission Electron Microscope (TEM) or calculated by an empirical formula. The value of C_v will be discussed in Section 3. According to the initial radius (~ 3.0 nm) and the concentration of void setting in the RT model, the S_w at the $D_{i,RT}$ can be obtained and is about 0.1%. R_s is the approximate irradiation swelling rate during the steady-state period of structural steel. In this work, R_s was set as 1%/dpa and 0.2%/dpa for the austenitic steels and F/M steels, respectively. The swelling behaviors in the dose range from the $D_{i,RT}$ to the target dose (after the incubation period), which can be simulated using the RT. Compared with the previous model, in which the initial dose used in the calculation is zero, the modification of introducing ML to predict the D_{onset} and obtain the $D_{i,RT}$ could improve the accuracy and applicability of the model.

2.2. Rate Theory of the Swelling Behavior after Incubation Period

Each incident neutron creates a series of knock-on atomic collisions in the reactor materials, creating point defects such as self-interstitial atoms (SIAs) and vacancies. After the cascade process, the surviving point defects would gather and form clusters including interstitial loops, vacancy loops, voids, and other microstructures. The flux of point

defect into interstitial dislocation loop is $Z_{v,i}D_{v,i}c_{v,i}$, where the subscript v and i denotes vacancy or interstitial atom, D means diffusion coefficient which can be acquired by $D = D_0 \exp(E_m/k_B T)$, c is the defect concentration [14]. The change rate of the radius of the interstitial loop is described as:

$$\frac{dr_{il}}{d\Delta} = \left(\frac{1}{Kb}\right) [Z_i D_i c_i - Z_v D_v c_v + Z_v D_v \bar{c}_{il}(\sigma)], \quad (3)$$

where Δ is the total radiation dose after time t , $\Delta = K \times t$, and K is the dose rate. The first and second terms of Equation (3) represent the bias term of the interstitial loop for trapping vacancies and SIAs. The last term is the atoms emitted thermally by the interstitial loops. $\bar{c}_{il}$ can be calculated by:

$$\bar{c}_{il}(\sigma) = c_v^e \exp\left\{-\frac{[(\gamma_{sf} + F_{el}(r_{il}))\Omega/b]}{k_B T}\right\}, \quad (4)$$

where c_v^e is the equilibrium concentration of vacancies. The energy term of the interstitial loop is given by $\gamma_{sf} + F_{el}(r_{il})$, where γ_{sf} is the stacking fault energy; in this rate model, γ_{sf} was set to 6.24×10^{22} (eV/cm²) for the F/M steel, while γ_{sf} was 9.4×10^{12} (eV/cm²) for the austenitic steel. F_{el} is the elastic energy and changes with the radius of the interstitial loop. F_{el} can be described as:

$$F_{el} = \frac{\mu b^2}{(1-\nu)(r+b)4\pi} \ln\left(\frac{r_{il}+b}{b}\right), \quad (5)$$

where ν is the Poisson's ratio and μ is the shear modulus. Similarly, the vacancy loop evolution under irradiation conditions can be described as:

$$\frac{dr_{vl}}{d\Delta} = \left(\frac{1}{Kb}\right) \left[Z_v D_v c_v - Z_i D_i c_i + Z_v D_v c_v^e \exp\left\{-\frac{[(\gamma_{sf} + F_{el}(r_{vl}))\Omega/b]}{k_B T}\right\} \right]. \quad (6)$$

Besides the interstitial loops, the third term of Equation (6) contains the change rate of the vacancy loop, accepting the vacancies emitted by other loops. The void radius may readily be shown as:

$$\frac{dr_v}{d\Delta} = \frac{1}{Kr_v} \left\{ D_v c_v - D_i c_i - D_v c_v^e \exp\left(\frac{2\gamma}{r_v} - p_g\right) \frac{b^3}{k_B T} \right\}, \quad (7)$$

where p_g is:

$$p_g = \frac{3n_g k_B T}{4\pi \left(r_v^3 - \frac{3b_v n_g}{4\pi} \right)}, \quad (8)$$

for simulating the gas generated during the neutron irradiation process. In this work, the dislocation loops and voids are considered to be the microstructure that would change with the accumulated doses.

To calculate the swelling rate under irradiation conditions, the concentrations of vacancies and SIAs are needed. The evolution equations of defects under neutron irradiation are described as:

$$\frac{dc_v}{dt} = \varepsilon K - \kappa D_v c_v D_i c_i - D_v \sum_j S^j Z_v^j (c_v - c_v^a), \quad (9)$$

$$\frac{dc_i}{dt} = \varepsilon K - \kappa D_v c_v D_i c_i - D_i \sum_j S^j Z_i^j c_i. \quad (10)$$

Since the point defect evolution can be assumed to reach a quasi-steady state a while after the cascade process, the steady-state equations of the concentrations of vacancies and SIAs are expressed as:

$$\varepsilon K - \kappa D_v c_v D_i c_i - D_v \sum_j S^j Z_v^j (c_v - c_v^a) = 0, \quad (11)$$

$$\varepsilon K - \kappa D_v c_v D_i c_i - D_i \sum_j S^j Z_i^j c_i = 0. \quad (12)$$

K is the production rate of point defects under irradiation, ε is the fraction of point defects that escape recombination during the cascade process [15]. The cascade efficiency ε depends on the primary recoil energy and irradiation type (electron, ion, or neutron irradiation). εK is the final surviving number of point defects after the cascade. Only these point defects can participate in the microstructure's evolution. We used 0.2 and 0.25 for the values of ε in austenitic steel and F/M steel under neutron irradiation, respectively [16]. κ is the recombination coefficient of the point defects, where $\kappa = \frac{4\pi R_c}{\Omega} \left(\frac{1}{D_v} + \frac{1}{D_i} \right)$, R_c is the recombination radius and D_v , D_i are the diffusion coefficients for vacancies and SIAs, respectively. S^j is the strength of the sink for type j .

In this work, the sinks considered in the model include network dislocations, dislocation loops, voids, and grain boundaries. The contribution of vacancy loops to the total sink strength is neglected since the latest research found that the loops are formed as part of the interstitial type, mainly under neutron irradiation [17]. Point defects can be captured by different types of dislocation and the sink strength is used to describe this capacity. The total sink strength for the capture of interstitials and vacancies are:

$$s_i^2 = s_{id}^2 + s_{iv}^2 + s_{ig}^2, \quad (13)$$

where s_{id}^2 , s_{iv}^2 , s_{ig}^2 are the strengths for the capture of the SIAs of dislocations (both network dislocations and dislocation loops), voids, and grain boundaries, respectively. The sink strengths for the capture of vacancies by different sinks are similar. These sink strengths have the following form:

$$s_{id}^2 = Z_{id} \rho_d, \quad (14)$$

$$s_{vd}^2 = Z_{vd} \rho_d, \quad (15)$$

$$s_{iv}^2 = s_{vv}^2 = 4\pi r_v C_v, \quad (16)$$

$$s_{ig}^2 = s_{vg}^2 = 24/d^2. \quad (17)$$

The void is considered as a neutral sink, so $s_{iv}^2 = s_{vv}^2$. Z_{id} is the average bias of dislocation loops and network dislocations for the capture of interstitials. Total dislocation density is calculated as:

$$\rho_d = \rho_d^0 + \rho_d^{il} + \rho_d^{vl}, \quad (18)$$

where ρ_d^0 is the density of the original network dislocations. The density of interstitial dislocation loops ρ_d^{il} is calculated as:

$$\rho_d^{il} = 2\pi r_{il} N_l, \quad (19)$$

where r_{il} is the interstitial loop radius. N_l is the total density of the interstitial loops [3]. The value of N_l can be either taken from experiments or from an empirical formula [9]. In this work, the applied stress is ignored.

The concentrations c_i and c_v can be solved using Equations (11) and (12). The sink strength of the different sinks can be obtained from Equations (13)–(19). During the irradiation process, the sizes of the dislocation loops and voids evolve with time, while the sizes of other sinks, such as network dislocations and grain boundaries, are treated

as constant. Due to the fact that sinks such as dislocation loops and voids change with the accumulated doses, while these sinks also influence the static point defect concentration, Equations (11)–(19) should be solved with Equations (3)–(7) together. Compared with the rate model used in previous research, in which the radii of voids and loops are the experimental data at a single point rather than the theoretical results [12], the improved RT model could predict the relationship between swelling and dose in a wide dose range due to the evolution of the microstructure thus described. The prediction capability of this rate theory model is also improved by considering the evolution of all the sinks. The initial conditions ($\Delta = D_{i,RT}$) for the order differential equations are: $r_v(D_{i,RT}) = 3 \times 10^{-7}(\text{cm})$, $r_l(D_{i,RT}) = 1 \times 10^{-7}(\text{cm})$, $p_g(D_{i,RT}) = \frac{2\gamma}{r_v(D_{i,RT})}$.

3. Results

Based on the ML-RT model, the swelling behavior in the dose range from 0 to hundreds of dpa can be simulated. To simulate the incubation period, the D_{onset} should be calculated first; to obtain the swelling performance after the incubation period, the rate equation should be solved through the initial conditions ($\Delta = D_{i,RT}$), as mentioned in Section 2.1. The main factors influencing the swelling in the ML-RT model are defect diffusion coefficients, dislocation densities, cascade efficiencies, etc. The corresponding parameters need to be used for the different types of steel.

3.1. Prediction of the Onset Dose of Swelling

The D_{onset} was predicted by using the ML based on the datasets of previous irradiation experiments. Figure 1 shows the results of mean absolute error (MAE) of the D_{onset} using each ensemble method that is mentioned in Section 2.1. Ideally, the MAE of the training set and testing set need to be as small as possible. From the results, RFR and GBR give better predictions since their MAEs of both the training and testing sets are lower than 10. In order to further verify the accuracy of the prediction, the RMSE and the R of different ensemble methods on the testing set were calculated, as shown in Figure 2. Among these methods, RFR appears to be the best choice, where the RMSE and R are both optimal. Figure 3 shows the optimal results of the predicted values of D_{onset} using the RFR method; the experimental data are also shown in the picture. The optimal values of R for both the training and testing sets are more than 0.9, while the RMSE for the test set is 9.22, indicating the good predictability of the ML method. Based on the RFR ensemble method and the dataset, the D_{onset} of the different materials under different irradiation conditions can be predicted easily. The results of the D_{onset} of each structural material would show in the following parts.

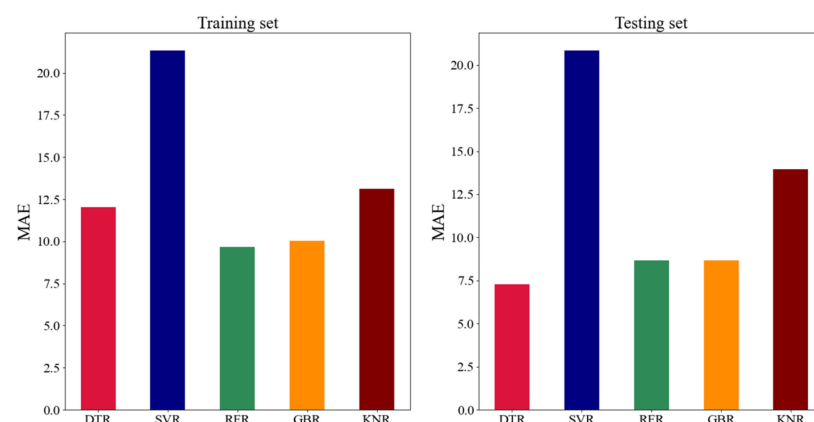


Figure 1. The mean absolute error of the different ensemble methods (decision tree regression (DTR), support vector regression (SVR), random forest regression (RFR), gradient boosting regression (GBR), and k-nearest neighbor regression (KNR)), training set, and testing set.

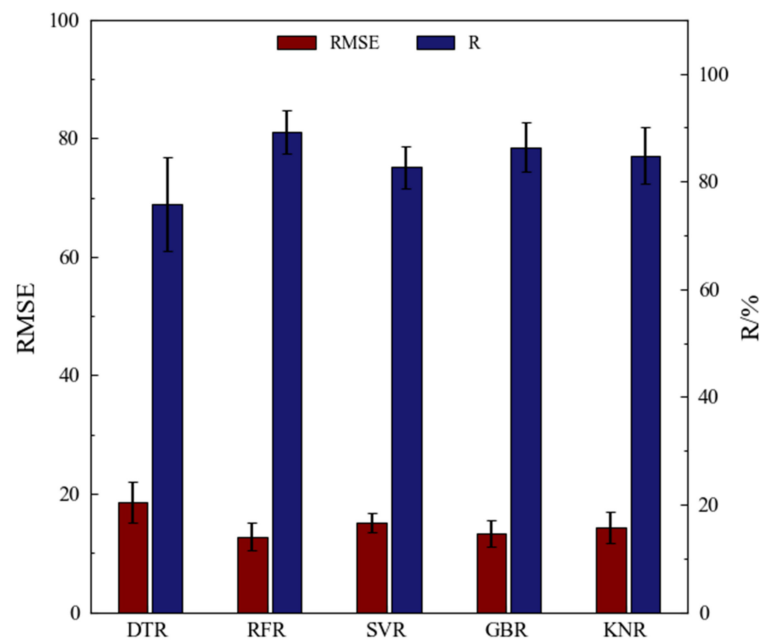


Figure 2. The correlation coefficient (R) and the root mean square error (RMSE), using different ensemble methods on the testing set.

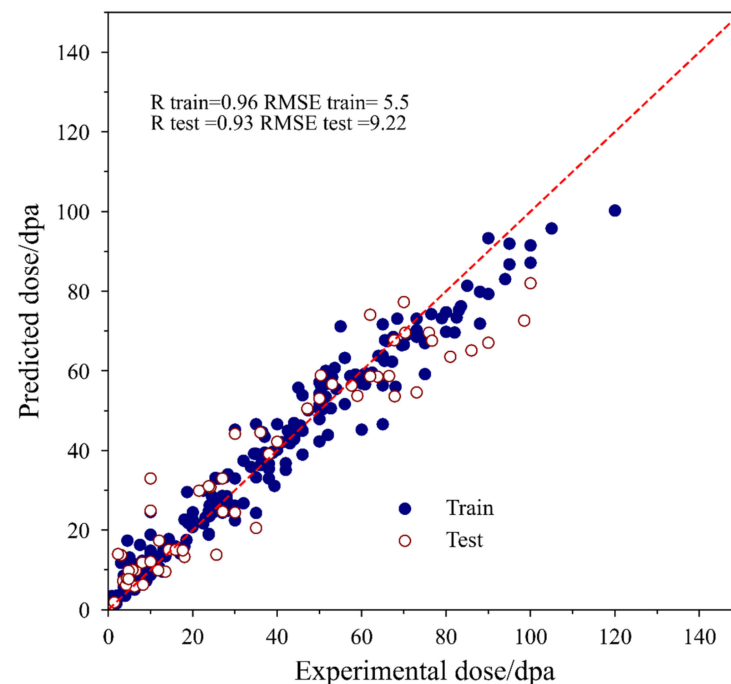


Figure 3. Comparison of the predicted and experimental results for the onset dose of swelling using the RFR method.

3.2. Simulation of Irradiation Swelling

3.2.1. Irradiation Swelling of Austenitic Steels

In the past few decades, a significant amount of experimental data on the irradiation swelling of austenitic steels, such as AISI 316, were obtained experimentally [18–20]. Thus, the swelling of AISI 316 was first used to verify the ML-RT model. The parameters of AISI 316 used in the calculation are shown in Table 2. The formation energies and migration energies of vacancies and SIAs are from the first-principle calculation and molecular dynamics (MD) simulation and are also verified by experimental data [21]. The value of the

recombination radius of the point defects was calculated to be 2–3.5 times that of the lattice constant [22], while the recombination radius was supposed to be as large as 7 times that of the lattice constant in a stable configuration [12]. Thus, the recombination radius in the range of from 2.5 to 7 times the lattice constant was tested in this work. The results indicate that the recombination radius has a limited influence on the evolution of the microstructure. As mentioned in Section 2, the voids and grain boundaries are neutral sinks, their values of sink bias are both 1.0 in terms of SIAs and vacancies. For non-neutral sinks, the bias values of the dislocation loops and network dislocations for SIA are 1.20. n_g is the number of gas atoms in the void, to simulate the neutron irradiation damage in the process of bubble formation [23]; here, the value of n_g was set to be 50 for the low helium production rate in the fission reactor. Other material parameters are similar to those used for AISI 316 in a previous work [24]. Table 3 summarizes the neutron irradiation parameters of AISI 316.

Table 2. Material parameters for austenitic AISI 316 and F/M Fe-9Cr binary alloy.

Material Parameters	AISI 316	Fe-9Cr
V-formation energy (eV)	1.8 [21]	1.9 [25]
SIA-formation energy (eV)	1.8 [21]	4.1 [25]
V-migration energy (eV)	1.4 [24]	1.1 [26]
SIA-migration energy (eV)	0.85 [24]	0.2 [26]
Dv0 (cm ² ·s ^{−1})	1.29×10^{-2} [21]	4.5×10^{-3} [25]
Di0 (cm ² ·s ^{−1})	1.29×10^{-2} [21]	3.0×10^{-5} [25]
Recombination coefficient	5.69×10^{26}	5.48×10^{27}
Recombination radius (cm)	1.27×10^{-7}	1.1×10^{-7}
Dislocation density (cm ^{−2})	1.5×10^{10} [27]	1.1×10^{11} [27]
Dislocation bias Z_{id}	1.20 [24]	1.05 [28]
Loop bias Z_{il}	1.20 [24]	1.05 [28]
Loop initial radius (cm)	10^{-7}	10^{-7}
Burger's vector (cm)	2.0×10^{-8}	2.86×10^{-8}
Lattice parameter (cm)	3.64×10^{-8}	2.8×10^{-8}
Poisson's ratio	0.264	0.3

Table 3. Neutron irradiation parameters of AISI 316, Fe-9Cr, JLF-1, and CLAM.

Parameters	AISI 316 [29]	Fe-9Cr [30]	JLF-1 [31]	CLAM [32,33]
Dose rate (dpa/s)	10^{-6}	10^{-6}	10^{-6}	10^{-6}
Cascade efficiency	0.2	0.25	0.25	0.25
Interstitial loop density (cm ^{−3})	$2.2 \times 10^4 \exp(1.7/k_B T)$	6.5×10^{16} at 425 °C	$1.8 \times 10^3 \exp(1.8/k_B T)$	1.1×10^{16}
Void concentration (cm ^{−3})	$3.0 \times 10^{16} \exp\{-[1/(k_B T \times 1.2)]^2\}$	8.7×10^{15} at 425 °C	7.45×10^{13} at 390 °C 2.2×10^{14} at 430 °C	5.0×10^{14}
Irradiation type	neutron	neutron	neutron	neutron
Neutron fluence (n m ^{−2})	10×10^{26} – 25×10^{26}	19×10^{26}	5.8×10^{26}	7.7×10^{26}
T (°C)	427–593	420	390–460	400

Based on the RFR method, the D_{onset} of AISI 316 under neutron irradiation in the temperature range from 450 °C to 590 °C was predicted to be 42–62 dpa. Figure 4a shows the swelling of AISI 316 steel in the temperature range of 450–590 °C after an irradiation dose of 120 dpa. The solid red curve represents the calculated results, while the black squares represent the experimental values [29]. As shown in Figure 4a, the swelling peak temperature of AISI 316 steel under neutron irradiation is predicted to be about 510 °C, which is close to the experimental data [29]. Because the swelling values at different temperatures were under different irradiation doses in the experiments, the dose value is labeled for each data point in Figure 4a. Besides the diffusion coefficient, equilibrium vacancy concentration, and thermal emission term, the concentrations of voids and interstitial loops also change according to temperature. In the temperature range of 300–600 °C, the interstitial loop concentration N_l is in the range of 7.5×10^{14} – 1.8×10^{16} cm^{−3} and the void

concentration C_v is in the range of $3.1 \times 10^{14} \sim 2.8 \times 10^{16} \text{ cm}^{-3}$. This data range is more close to the experimental results [34] compared with the values in the previous rate theory [3]. The formulas of N_l and C_v , with their temperatures, are shown in Table 3. The results of the swelling rates are different at different temperatures; for example, they are about 0.4%/dpa at 427 °C and about 0.9%/dpa at 510 °C. The major reason that influences the swelling rate under different temperatures is the diffusion coefficient of the point defects. The swelling could not be simulated properly at high temperatures using the previous rate theory model [2]; the main reasons for this are the improper description of the vacancy loop evolution and cascade effects, as the point defect flux $Z_{i,v}D_{i,v}c_{i,v}$ at high temperatures would cause the accumulating rate to become negative. In this work, the effect of the vacancy loop has been ignored while the focus is on the evolution of interstitial loops. The cascade efficiency of point defects production has been used to calculate the value of the surviving point defects after the cascade process. By using this modified ML-RT model, the neutron swelling at high temperatures ($>525^\circ\text{C}$) can be predicted, as shown in Figure 4a.

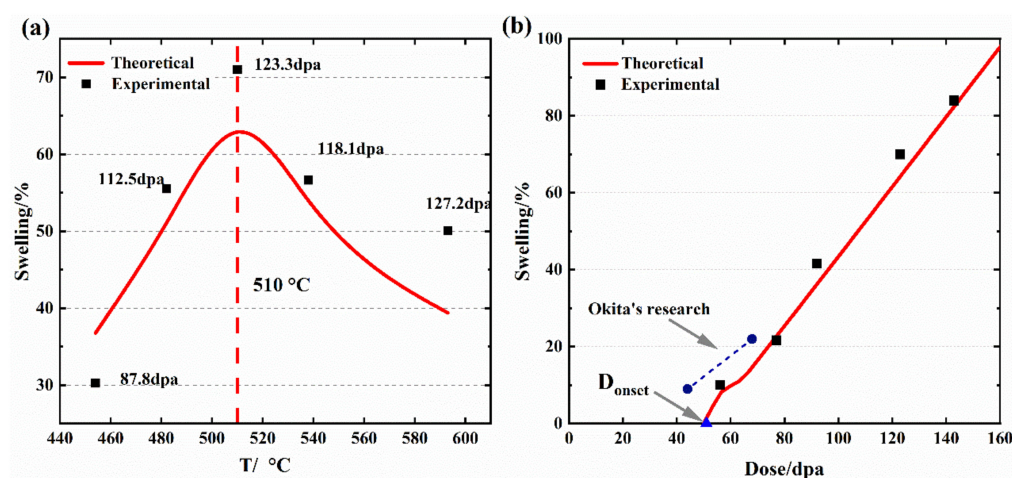


Figure 4. Neutron irradiation swelling of austenitic AISI 316 steel: (a) swelling as a function of temperature with the irradiation dose of 120 dpa, (b) swelling as a function of the dose at 510 °C, where the black squares represent experimental data from [29] and the red line corresponds to the results of theoretical calculations.

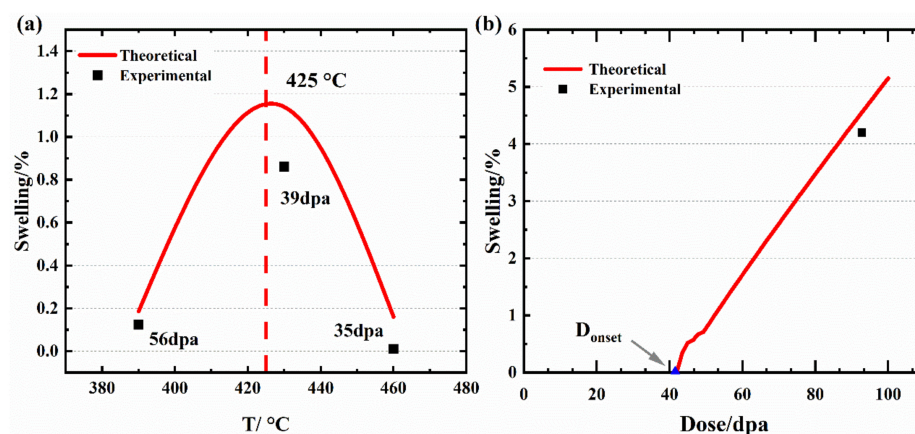
Figure 4b shows the theoretical and experimental results of swelling in AISI 316 steel at 510 °C. We chose a temperature of 510 °C because this is the irradiation peak temperature and there are enough experimental swelling data. The black squares represent the neutron irradiation experimental data in AISI 316, which are listed in Table 4 [29]. The blue point represents the D_{onset} predicted by ML, which is about 53.6 dpa for 316 at 510 °C. The $D_{i,\text{RT}}$ could then be obtained, which is 53.7 dpa. As shown in Figure 4b, the swelling rate during the steady-state period (53.7~180 dpa) is calculated to be about 0.9%/dpa, based on the RT model, which is in good agreement with the experimental results [20] and with previous theoretical results [12]. Compared with Okita's work, which could only predict the swelling of a single point, as marked in Figure 4b, our work could predict the swelling evolution with accumulated doses. In this period, the swelling rate is influenced by the combined effect of neutral sinks and non-neutral sinks. Obviously, if neutral sinks such as voids increase more quickly than non-neutral sinks such as loops, the swelling rate will become slower. In the theoretical results, the steady-state swelling rate does not change significantly, indicating that the impact of voids and loops on swelling are close to each other. Compared with the classic RT model, the coupled ML-RT model can predict swelling behaviors more appropriately, especially for the onset dose. The validity of the model is verified by the comparison between the theoretical and experimental results up to 150 dpa [29], as shown in Figure 4a,b.

Table 4. Experimental data of neutron swelling in austenitic AISI 316 steel under different irradiation doses or at different temperatures from [29].

Dose/(dpa)	Swelling/% 510 °C	T/(°C)	Dose/(dpa)	Swelling/%
123.3	71.1	427	87.8	16.9
141.3	87.5	482	112.5	46.4
89.7	41.5	510	123.3	51.6
71.8	23.6	538	118.1	41.5
56.1	11.5	593	127.2	29.6

3.2.2. Irradiation Swelling of F/M Steels

To simulate the swelling of F/M steels, the ML-RT model is similar to that used for austenitic steels in Section 2.2, while the parameters for F/M steels are taken (listed in Tables 2 and 3). The swelling behaviors under neutron irradiation in the F/M steels JLF-1 and Fe-9Cr binary alloy are shown in Figure 5a,b. The red solid curve represents the calculated results while the black squares represent the experimental data [31,35]. The parameters of the F/M steels are listed in Tables 2 and 3. As shown in Figure 5a, based on the ML-RT model, the swelling peak temperature of JLF-1 under neutron irradiation is predicted to be about 425 °C with an irradiation dose of 60 dpa in the temperature range of 380–470 °C, which is close to the swelling peak temperature of 420 °C garnered from neutron irradiation experiments with JLF-1 [31]. In the calculating of the RT of JLF-1, all the parameters are from experimental data and the literature [26,31]. The D_{onset} of JLF-1 in this temperature range, as predicted by ML, is 36–47 dpa and, in the steady-state period, the swelling rate is about 0.07%/dpa at 430 °C and about 0.03%/dpa at 390 °C according to the model. The concentration of interstitial loops varies as a function of temperature, as shown in Table 3. The concentrations of voids are based on the statistical results from previous experiments [31].

**Figure 5.** Neutron irradiation swelling of F/M steels: (a) swelling as a function of temperature in JLF-1 with an irradiation dose of 60 dpa, (b) swelling as a function of the dose at 425 °C in Fe-9Cr binary alloy where the black squares represent experimental data from [29,31].

As shown in Figure 5b, the relationship between the swelling and doses of the F/M steel and Fe-9Cr binary alloy was predicted by the ML-RT model in the dose range from 0 to 100 dpa at 425 °C. The swelling rate at a steady state is calculated to be about 0.08%/dpa, which is much lower than that of AISI 316 at peak temperature. The D_{onset} of Fe-9Cr, based on ML, is predicted to be 42 dpa at this temperature. Compared with that of AISI 316, which is about 50 dpa, the value of D_{onset} of Fe-9Cr is even lower. The reason for this result will be discussed in Section 4. The theoretical results of swelling agree with the experimental data, which are about 4.1% at 91 dpa and 420 °C [30]. This agreement verifies the feasibility of using the material parameters of the F/M steels to simulate the swelling of corresponding steels by this coupled model, such as Fe-9Cr and JLF-1. When the accumulative doses

reach 100 dpa, the total swelling of Fe-9Cr becomes 5.2%, which is much lower than that of AISI 316. Both the theoretical results and experimental data confirm that the martensitic steels Fe-9Cr and JLF-1 have better swelling resistance than the austenitic AISI 316. From the ML-RT model, the lower steady-state swelling rate has a far greater impact than the difference in D_{onset} of austenitic and F/M steels.

3.2.3. Prediction of Irradiation Swelling in CLAM Steel

The verified coupled ML-RT model can be used to predict the swelling behaviors of some potential nuclear structural materials under a wide range of irradiation doses. As one of the candidate RAFM steels for fusion structural materials, China low-activation martensitic (CLAM) steel is chosen for simulation in this work. As the candidate reactor's structural material, the neutron swelling behavior of CLAM steel is lacking in high-dose conditions. The calculating model developed in this work gives a feasible method by which to predict swelling behaviors in the dose range from 0 to hundreds of dpa. According to the composition and material parameters of CLAM steel, the predicted results are shown in Figure 6, where swelling behavior with a dose up to 180 dpa at 400 °C has been predicted. The temperature was chosen to be 400 °C in the calculation because this value is close to the peak temperature of 425 °C of F/M steels and this is the irradiation temperature of the chosen experimental data under neutron irradiation. The cascade efficiency is also set as 0.25 in the calculations. Considering that the performance of CLAM steel after fission neutron irradiation was similar to that of F82H when irradiated under similar irradiation conditions [36], the parameters of CLAM steel in Equation (18) are of the same order of magnitude as those of F82H in the neutron irradiation experiments [32,33]. As in the predicted results shown in Figure 6, the D_{onset} is 42 dpa of CLAM steel from ML and the value of $D_{i,RT}$ is 42.4 dpa. The swelling rate at 400 °C during the steady-state period is about 0.03%/dpa from the RT model. These theoretical results indicate the better swelling resistance of CLAM steel than those of AISI 316 and Fe-9Cr. Compared with the results of JLF steels, besides the irradiation temperature, the main reason for the difference in swelling behavior is the density of voids and dislocation loops, based on the experimental data that are listed in Table 3. When the accumulation doses are up to 180 dpa, the total swelling of CLAM steel is still below 3%, which means that the steel may meet the reactor design requirements in terms of irradiation swelling resistance. This predicted result is close to the experimental swelling data for Fe-9Cr and another RAFM steel, F82H, in HFIR [32] at about 400 °C.

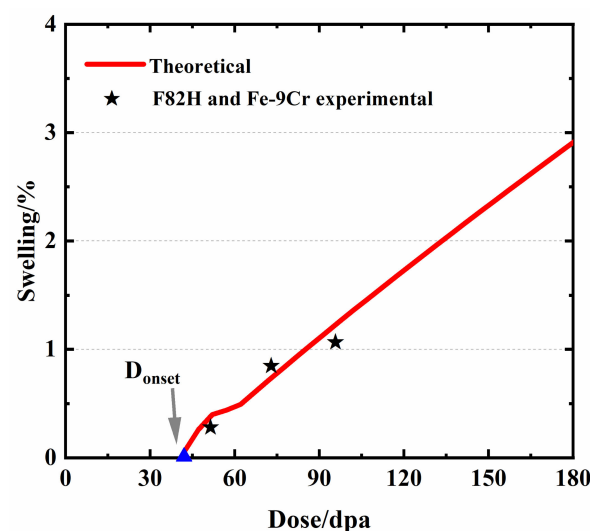


Figure 6. Predicted irradiation swelling of CLAM steel as a function of the dose under neutron irradiation (with low helium production rate in fission reactor) at 400 °C and compared with the experimental data of F/M steels, data from [32,33]. Due to the production of H and He in the fusion reactor, the swelling in the fusion reactor may be higher than in the predictions.

4. Discussion

As we know, F/M steels have better swelling resistance compared with austenitic steel. The possible reasons for this phenomenon are complicated [37]. In this work, we try to analyze the major factors, based on the ML-RT model calculation and the results shown in Section 3. The swelling performance depends on D_{onset} , cascade efficiency, defect diffusion coefficient, sink strength, etc. We mainly focus on these factors.

4.1. Steady-State Swelling Onset Dose

The D_{onset} value could influence the swelling behavior strongly. There are two different viewpoints regarding the D_{onset} of F/M and austenitic steels. The first viewpoint is based on the research of Garner et al. The F/M steels exhibit a longer swelling incubation period and have a higher D_{onset} than that of austenitic steels, and several experiments may validate these claims [29]. For example, a longer swelling incubation period has been observed under the neutron irradiation of Fe-9Cr and HT-9 in Fast Flux Test Facility (FFTF). The D_{onset} of HT-9 at 425 °C in FFTF is more than 100 dpa [29]. Another viewpoint is from the research of T. Morimura et al.; the authors believed that the D_{onset} of JLF series steels is about 30 dpa, based on the irradiation experiments in FFTF [31]. In addition, several experiments also indicate that the D_{onset} is about 25 dpa in the Experimental Breeder Reactor-II (EBR-II) of Fe-Cr alloy [38], which is even lower than that of austenitic steels. In general, the D_{onset} of F/M steels is not always much higher than that of austenitic steels, based on numerous irradiation experiments; many factors, such as reactor types, temperature, irradiation rate, etc., could influence the results. The research of Garner et al. showed a comparison of the swelling of Fe-Cr alloys in EBR-II and FFTF-MOTA; Fe-Cr alloys exhibit a longer swelling incubation period in FFTF, where the D_{onset} is about 100 dpa, while in EBR-II, the D_{onset} is only about 25 dpa [29].

In this work, the dataset used in ML contains these different values of D_{onset} from irradiation experiments and gives the predicted results for each steel based on these experimental data. Like the results shown in Section 3, the predicted D_{onset} of F/M steels is not significantly higher than that of austenitic steels. The reason is that numerous neutron irradiation experiments have indicated that the onset of F/M steels is close to the value of austenitic steels. Therefore, based on the simulated results, we prefer the view that the D_{onset} of F/M steels may not be significantly higher than that of austenitic steels. The swelling rate after the incubation period should be the most important factor to impact the different swelling behaviors in the two types of steels.

4.2. Cascade Efficiency

The cascade efficiency is strongly associated with recoil energy and irradiation type. The values of cascade efficiency in this work are about 0.2 for austenitic steels and 0.25 for F/M steels, respectively, based on previous research [16]. A higher value of cascade efficiency means that more point defects could participate in the evolution of defect clusters and lead to swelling. In this ML-RT model, different values of cascade efficiency cause about 5% lower levels of swelling of the F/M steels after 180 dpa than those of the austenitic steels.

4.3. Point Defect Diffusion

The migration of point defects (vacancy, SIA) in materials can influence the growth of voids under irradiation. In previous research [39], the disparity of the diffusion capacity between vacancy and SIA was considered to cause different swelling behaviors in different materials under irradiation. Interstitial atoms move much faster than vacancies under irradiation, which results in the phenomenon that the same types of point defects have more likelihood of gathering and generating interstitial clusters or voids. The vacancy migration energy is 1.3 eV–1.4 eV in the fcc steels, while it is about 0.65 eV in the bcc steels [25]. Considering the fact that a large proportion of the F/M steels used as nuclear structural materials have adulteration elements, the vacancy migration energy rises up to 1.2 eV in RAFM steels [40]. The migration energies of SIAs in the austenitic steels and the

F/M steels are about 0.85 eV and 0.2 eV, respectively. To study the influence of point defect diffusion on swelling, the vacancy and SIA diffusion coefficients at different temperatures are calculated and are shown in Figure 7. Figure 7a shows the diffusion coefficients of vacancy for each temperature of F/M and austenitic steels, while (b) is that of SIA. The gap of diffusion coefficients between the SIA and vacancy is larger in the austenitic steels than the F/M steels. This means that it is easier to form voids or clusters in the austenitic steels, which have a higher swelling rate.

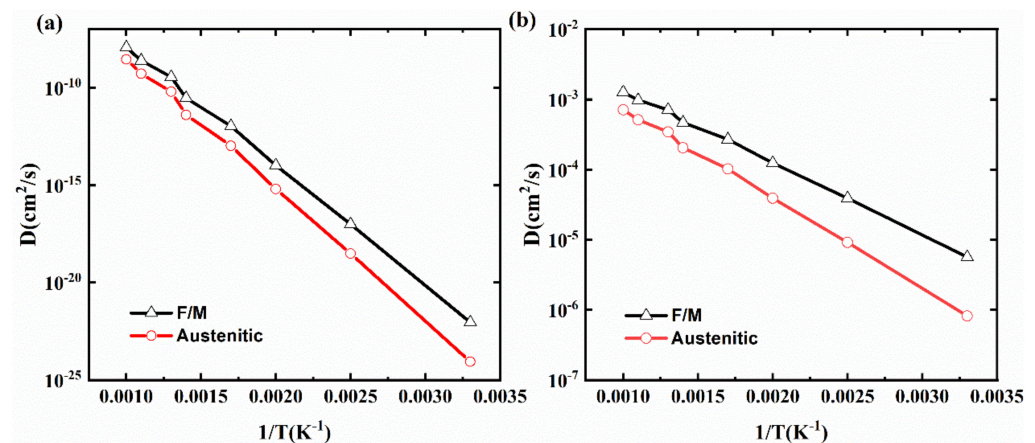


Figure 7. (a) The diffusion coefficients of vacancy, (b) the diffusion coefficients of SIA in F/M steels and austenitic steels under different temperatures.

Figure 8a shows the swelling results with different diffusion coefficients and diffusion gaps between the vacancy and interstitial areas using the MT-RT model. The migration energies of the vacancy and SIA are set as 1.4 eV and 0.85 eV in AISI 316, respectively, and the swelling plots are shown as a black line. To study the influence of different diffusion coefficients, the value of the migration energy of vacancy is set as 1.2 eV, while the migration energy of SIA stays the same and the results are plotted as a blue line. For another condition, the migration energy of the vacancy is still 1.4 eV, while the migration energy of SIA is set as 1.4 eV for testing; the results are plotted as a red line. As shown in Figure 8a, compared with the swelling results of AISI 316 (black line), the swelling at 180 dpa is about 9% lower, as caused by the diffusion gap, and 15% lower from the difference in the migration energies of vacancy.

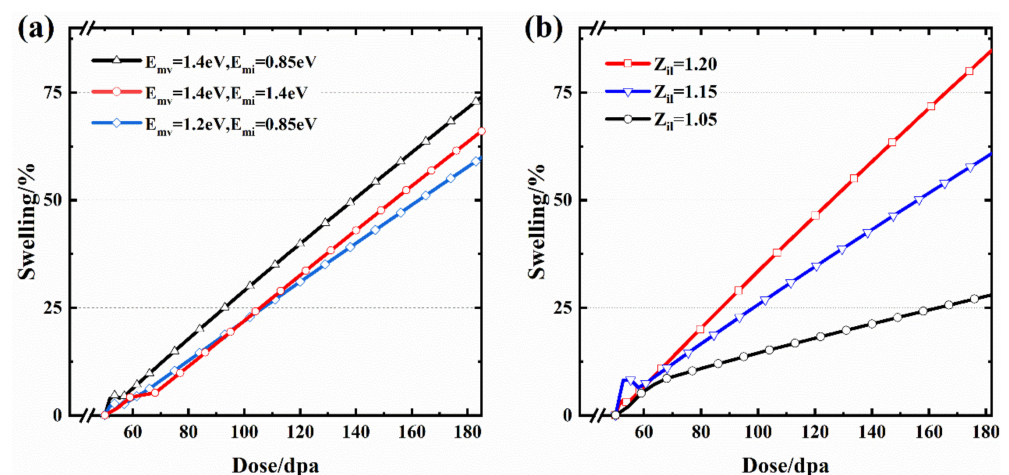


Figure 8. (a) The swelling of AISI 316 with different E_{mv} or E_{mi} . The black line with triangles: $E_{mv} = 1.4 \text{ eV}$ and $E_{mi} = 0.85 \text{ eV}$; the red line with circles: $E_{mv} = 1.4 \text{ eV}$ and $E_{mi} = 1.4 \text{ eV}$; the blue line with squares: $E_{mv} = 1.2 \text{ eV}$ and $E_{mi} = 0.85 \text{ eV}$. (b) The swelling plots as a function of doses at 450 °C with the loop biases Z_{il} equal to 1.05, 1.15, and 1.20, respectively.

4.4. Sink Strength

The sink strength depends on the bias and the density of different types of sinks, including network dislocation, dislocation loops, voids, grain boundaries, etc. From previous irradiation experiments, the densities of sinks could be acquired in most cases. In this work, we compared the influence of the sink density for different materials. For example, the network dislocation densities are $1.5 \times 10^{10} \text{ cm}^{-2}$ and $1.1 \times 10^{11} \text{ cm}^{-2}$ and the dislocation loop densities are $2.5 \times 10^{15} \text{ cm}^{-2}$ and $4.5 \times 10^{15} \text{ cm}^{-2}$ for 316 and Fe-9Cr, respectively [27]. This density difference results in about 3% lower swelling in Fe-9Cr than that in AISI 316. In general, the more non-neutral sinks there are, the greater the amount of swelling the material has. However, this density discrepancy of dislocations in AISI 316 and Fe-9Cr is not sufficient to cause the much lower swelling seen in Fe-9Cr.

The bias of sinks influences the swelling in different types of steels with fcc and bcc crystal structures [41]. The bias leads to the generation of excessive vacancies, which affect the growth rate of voids in the material. As we know, void and grain boundaries are neutral sinks in which the vacancy and SIA can be absorbed equally. In non-neutral sinks, such as dislocation loops and network dislocation, SIA can be absorbed more easily than a vacancy. The bias leads to a surplus of vacancies under irradiation and results in void formation. To demonstrate the influence of bias on swelling, the swelling curves of AISI 316 with different values for the loop bias, are plotted in Figure 8b. The swellings at 180 dpa are 23%, 59%, and 81% when Z_{il} equals 1.05, 1.15, and 1.20, respectively. The results clearly show that bias is the dominant factor causing the different swelling behaviors between austenitic steels and F/M steels.

4.5. Helium Effects

In this work, the experimental data for swelling were mainly conducted in fission reactors with a low He production rate. The high generation of helium in a fusion reactor would influence the swelling performance strongly. The irradiation research of Ni-doped specimens of RAFM steel and the irradiation experiments in a spallation neutron source all indicate the influence of bubble swelling [32,42,43]. However, due to the swelling data in fusion reactors being lacking, predictions of the swelling in a fusion reactor with a high He production rate are difficult to verify. The model developed in this manuscript focuses on void swelling since the experimental data were mainly from neutron irradiation in fission reactors with a low He production rate (e.g., EBR-II, FFTF, High Flux Isotope Reactor (HFIR)).

In general, predicting the swelling behavior of CLAM steel in a fusion reactor environment requires the consideration of helium effects (bubble swelling) and void swelling. This model lays the foundation for that consideration, and Equation (8) gives a simplified description of the gas atoms that are induced by irradiation with a very low He concentration (about 1 appm). For the high generation of helium conditions, such as the fusion reactor and spallation neutron source, the generation term of a helium bubble is needed, and the temperature effects also need re-assessment. In our next work, we would try to extend this model to simulate both void swelling and bubble swelling.

Compared with the previous rate theory model, this ML-RT model has been improved by introducing ML for predicting the D_{onset} and the $D_{i,\text{RT}}$; furthermore, the fitting parameters were reduced and more suitable values for sink strengths were employed. The evolutions of both the dislocation loop and void with cumulative doses were considered in this model and improved the model's predictive ability under a target dose without experimental data. The coupled ML-RT model has advantages in predicting the swelling behavior of materials under high-dose irradiation and can be applied to evaluate the void swelling behaviors of metallic structural materials in a future fusion reactor under high-dose neutron irradiation efficiently and effectively.

5. Conclusions

A coupled ML-RT model consisting of the ML method and an improved RT model was developed in this work. The D_{onset} could be predicted for different materials by using the ML method. Based on the RT model, the peak temperatures are about 510 °C and 425 °C for austenitic steels (e.g., AISI 316) and F/M steels (e.g., JLF-1), respectively. The swelling rates were calculated to be about 0.9%/dpa for 316 at 510 °C and 0.08%/dpa for Fe-9Cr at 425 °C, during their steady-state periods. In addition, the swelling of CLAM steel was predicted to be less than 3% under 180 dpa neutron irradiation at 400 °C. Based on this ML-RT model, the factors influencing the swelling of different materials are discussed and bias is found to be the dominant factor. These results will be helpful in the evaluation of the neutron irradiation swelling behavior of candidate structural materials serving in the reactors and can also provide useful guidance for the optimization of these structural materials.

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