Molecular dynamics simulation of displacement cascades in Ni-Mo alloy postprint

Authors: HU Neng-Wen, QI Mei-Ling, XIAO Shi-Fang, DENG Hui-Qiu, REN Cui-Lan, HU Wang-Yu

Date: 2023-06-18T00:00:00+00:00

Abstract

Molecular dynamics method is used to investigate the displacement cascades in Ni-Mo binary alloy. Effects of the irradiation temperature, energy of the primary knock-on atoms and concentration of solute Mo atoms are taken into consideration on radiation damage to the Ni-Mo alloy. It is found that Mo atoms reduce production of the Frenkel pairs at 100 K, while they enhance defect production at 300 K and 600 K. Size of the largest defect clusters decreases with increasing concentrations of Mo atoms (CMo) at 100 K, but it increases with CMo at 300 K and 600 K. Most of the point defects get clustered in cascades leaving only a few vacancies and interstitials isolated.

Full Text

Preamble

Molecular Dynamics Simulation of Displacement Cascades in Ni-Mo Alloy

HU Neng-Wen,[1],[2] *QI Mei-Ling,*[2] *XIAO Shi-Fang,*[3] *DENG Hui-Qiu,*[3],† *REN Cui-Lan,*[4] *and HU Wang-Yu*[1],[3],‡

[1]College of Materials Science and Engineering, Hunan University, Changsha 410082, China
[2]Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China
[3]Department of Applied Physics, School of Physics and Electronics, Hunan University, Changsha 410082, China
[4]Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201800, China

(Received April 22, 2015; accepted in revised form June 9, 2015; published online December 20, 2015)

Abstract: The molecular dynamics method is used to investigate displacement cascades in Ni-Mo binary alloy. Effects of irradiation temperature, energy of primary knock-on atoms, and concentration of solute Mo atoms on radiation damage to the Ni-Mo alloy are considered. It is found that Mo atoms reduce production of Frenkel pairs at 100 K, while they enhance defect production at 300 K and 600 K. The size of the largest defect clusters decreases with increasing concentrations of Mo atoms (C_{Mo}) at 100 K, but increases with C_{Mo} at 300 K and 600 K. Most point defects become clustered in cascades, leaving only a few vacancies and interstitials isolated.

Keywords: Molecular dynamics method, Displacement cascade, Ni-Mo alloy
DOI: 10.13538/j.1001-8042/nst.26.060603

Introduction

High-temperature molten salt reactors (MSRs) represent one of the most promising and safe types of advanced Generation-IV fission reactors [1, 2]. These reactors are expected to operate with fluoride salt melts as fuels and coolants at high temperatures, which imposes several stringent requirements on candidate structural materials, including high corrosion resistance to fluoride salt melts, good manufacturability, high-temperature strength, and sufficient radiation resistance [3]. Hastelloy, particularly Hastelloy N, with Ni and Mo as its predominant constituents, represents an important candidate for in-core structural materials in MSRs due to its high corrosion resistance to fluoride salt melts, high-temperature strength, and good manufacturability [4].

Nevertheless, bombardment by energetic particles induces elastic and inelastic collisions in materials, causing thermodynamic phenomena such as local heat spikes and radiation heat waves [5]. Available experimental results demonstrate that ions, electrons, and neutrons can cause significant damage to the microstructures of Hastelloy, forming point defects, dislocation loops, stacking fault tetrahedrons (SFTs), and precipitates, which leads to swelling, ductility loss, high-temperature strength degradation, hardening, embrittlement, and other detrimental effects [6-10]. These phenomena can severely limit the lifetime of MSRs. To improve the radiation resistance of nickel-based alloys, chemical composition optimization—currently used for commercial alloys—may prove an effective approach [11].

First-principles calculations have shown that solute atoms affect the cracking properties of Hastelloy N [12], underscoring the importance of studying solute atom effects on alloy radiation resistance. Cascade collisions occur on picosecond timescales, which are too rapid for experimental observation. Molecular dynamics methods have been widely employed to study displacement cascades since 1993, when they first succeeded in primary damage investigations [13], and remain a standard tool for examining primary defect behaviors in materials [14–16].

To the authors' knowledge, however, little attention has been paid to radiation damage in Ni-Mo alloys [17], particularly regarding displacement cascades in Hastelloy. Since Ni and Mo constitute the main components of Hastelloy N [4], this paper constructs simplified Ni-Mo alloy models to investigate the effects of irradiation temperature, primary knock-on atom (PKA) energy, and solute Mo atom concentration on displacement cascades.

Models and Methods

Molecular dynamics simulations of displacement cascades in Ni-Mo alloy are performed using modified MOLDY codes. A set of interatomic potential functions based on the modified analysis embedded atom method (MAEAM) are fitted to experimental and first-principles calculation results. A modified term is added to scale errors resulting from the spherical approximation of non-s-electrons. The total energy of a crystal (E_{tot}) with N atoms can be calculated by:

$$E_{tot} = \sum_i F_i(\rho_i) + \sum_{i,j,i\neq j} \phi(r_{i,j}) + \sum_i M_i(P_i)$$

where $\sum_i M_i(P_i)$ is the empirical modification term, $F(\rho)$ is the embedding term, and $\phi(r)$ is the repulsive term. The cross-section describing the interaction between Ni and Mo atoms can be described by $\phi_{AB}(r) = [\phi_{AA}(C_{1r}) + \phi_{BB}(C_{2r})]$, where μ, C_1, and C_2 are adjustable parameters. The formulae based on MAEAM theory can be found in our previous papers [18]. Information from the Ni-Mo binary alloy phase diagram is incorporated, with formation enthalpies of common stable phases (such as δ-NiMo, D0a and DO22 structural Ni3Mo, and D1a structural Ni4Mo) fitted into the cross-interaction potential parameters of Ni and Mo. The Mo solubility in Ni is considered and fitted approximately as 20 at.% atomic concentration, which agrees well with the Ni-Mo alloy phase diagram.

Ni-Mo binary alloy models with solute Mo atom concentrations (C_{Mo}) of 3.0 at.%–15 at.% are established, with Mo atoms randomly distributed at substitutional sites. All alloy models with dimensions of $20a_0 \times 20a_0 \times 20a_0$ are relaxed to equilibrium under the NPT ensemble for approximately 250 picoseconds to obtain lattice constants at each temperature and Mo concentration. All displacement cascade simulations are performed in larger boxes of $60a_0 \times 60a_0 \times 60a_0$ at temperatures of 100 K, 300 K, and 600 K under the NVE ensemble, where a_0 is the lattice constant corresponding to the temperature and C_{Mo}. PKA energies of 5 keV, 10 keV, 20 keV, 30 keV, and 40 keV are used. To avoid channeling effects, all PKAs are set along the $\langle 135 \rangle$ direction, one of the high-index directions. The time step is one femtosecond in general, with each cascade simulation running for 15,000 time steps (approximately 15 ps). The Wigner-Seitz cell method is adopted to identify defects in each cascade simulation [19]. An additional cutoff separation of $1.25a_0$ is used to identify defect clusters, such that two point defects belong to the same cluster if they are within $1.25a_0$ of each other. All defects generated in cascade collisions are quenched to 0 K before cluster analysis. Each cascade is run five times for improved statistics.

Results and Discussion

Dislocation loops greatly affect defect numbers, so defects within dislocation loops are excluded from statistical analysis of defect counts. The number of defects increases rapidly with PKA energy, as PKAs initialized with high kinetic energy cause severe collisions under the NVE ensemble, displacing numerous lattice atoms. Consequently, more defects remain in the system after cascade cooling, although most displaced atoms can recombine with vacancies.

Figure 1 shows that the number of defects resulting from cascade collisions decreases slightly with increasing Mo atomic concentration at 100 K, but increases rapidly with C_{Mo} at 300 K and 600 K. Mo atoms appear to play opposite roles in defect generation: they suppress defect formation at low temperatures but enhance it at higher temperatures. This can be understood as a synergistic effect of irradiation temperature and Mo atom concentration. Randomly distributed solute Mo atoms in Ni-Mo alloy can prevent atoms from being displaced and from recombining with vacancies during displacement cascade processes. Lattice atoms are more difficult to displace at low temperatures than at high temperatures because thermal vibrations are weaker in low-temperature environments. Consequently, low temperature combined with high Mo concentration inhibits defect production. At high temperatures, lattice atoms can be displaced more easily, and once displaced, recombination becomes difficult because Mo atoms prevent displaced atoms from recombining with vacancies, substantially increasing defect numbers.

In Fig. 1(b), the result at C_{Mo} = 3.0 at.% and PKA energy = 40 keV at 300 K is anomalous due to the formation of large dislocation loops in the stable state, where atoms aggregate into dislocation loops leaving an amorphous zone in the system. The distribution of cascade defects in Ni-Mo alloy is similar to that in iron and tungsten: irradiation-induced vacancies distribute in the cascade center while interstitials migrate to the periphery [20, 21].

Most defects generated in cascade processes distribute in clusters, and the number of small clusters increases with PKA energy. Further analysis of cluster distribution shows that most defect clusters are smaller than 10 defects, with the number of small clusters increasing with PKA energy. Typical results for cluster number distribution as functions of PKA energy and cluster size are shown in Fig. 2. The increasing number of small clusters occurs because high-energy PKAs generate high-temperature zones where interstitials and vacancies migrate and aggregate more easily. The clustering behavior of cascade defects in Ni-Mo alloy differs from that in Fe-W alloy, where interstitials typically occur as dumbbells, crowdions, or sometimes dislocation loops [22-24]. In Ni-Mo alloy, however, most interstitials cluster into SFTs or interstitial loops, with only a very small fraction remaining isolated.

The Mo concentration in Ni-Mo alloy also influences the size of the largest cluster. As shown in Fig. 3, the number of small clusters decreases slightly with increasing C_{Mo}. The size of the largest clusters in Ni-Mo alloy decreases with increasing Mo atomic concentration at 100 K, but increases sharply with Mo content at 600 K. This may be because the cascade simulation time is insufficient for vacancies to migrate and form clusters. Consequently, small cluster formation depends on interstitial atom aggregation rather than vacancy clustering. Adding more Mo atoms to Ni-Mo alloy reduces the total number of point defects at low temperatures, making interstitials unavailable for cluster formation, hence decreasing both the number of small clusters and the size of the largest clusters with increasing C_{Mo}. In high-temperature environments, Mo atoms increase defect numbers and most displaced atoms collapse into dislocation loops because the limited time prevents recombination. Solute Mo atoms at higher atomic fractions lead to more interstitial aggregation into loops, which also limits available interstitials and decreases the number of small clusters. Therefore, the number of small clusters decreases with increasing C_{Mo}, but the size of the largest clusters increases with C_{Mo}.

Conclusion

The effects of solute Mo atoms, irradiation temperature, and PKA energy on displacement cascades in Ni-Mo alloy are investigated using molecular dynamics. The conclusions are as follows: (1) Solute Mo atoms decrease the size of defect clusters. At 100 K, they reduce Frenkel pair production, while at 300 K and 600 K they enhance defect production. (2) Most point defects become clustered during cascade processes, with only a few vacancies and interstitials remaining isolated. Dislocation loops can form in cascades under high PKA energies and/or high temperatures. (3) Most vacancies generated in cascade processes remain in the cascade center, leading to formation of a depleted zone in the cascade region. A small fraction of single point defects and small clusters can form far from cascade centers.

References

[1] Mathieu L, Heuer D, Brissot R, et al. The thorium molten salt reactor: Moving on from the MSBR. Prog Nucl Energ, 2006, 48: 664-679. DOI: 10.1016/j.pnucene.2006.07.005
[2] Le Brun C. Molten salts and nuclear energy production. J Nucl Mater, 2007, 360: 1-5. DOI: 10.1016/j.jnucmat.2006.08.017
[3] Yvon P and Carré F. Structural materials challenges for advanced reactor systems. J Nucl Mater, 2009, 385: 217-222. DOI: 10.1016/j.jnucmat.2008.11.026

[4] Bakai A S. Combined effect of molten fluoride salt and irradiation on Ni-based alloys. Mater Issues Gen IV Systems, 2008, 537-557. DOI: 10.1007/978-1-4020-8422-5_{27}

[5] Ding Y N, Yao Z Y, Miao W Y, et al. Experimental observation of radiation heat waves. Nucl Sci Tech, 1997, 8: 43-45.

[6] Jin S X, Guo L P, Yang Z, et al. Microstructural evolution in nickel alloy C-276 after Ar+ ion irradiation. Nucl Instrum and Meth B, 2011, 269: 209-215. DOI: 10.1016/j.nimb.2010.12.004

[7] Jin S X, He X F, Li T C, et al. Microstructural evolution of P92 ferritic/martensitic steel under Ar+ ion irradiation at elevated temperature. Mater Charact, 2012, 68: 63-70. DOI: 10.1016/j.matchar.2012.03.009

[8] Jin S X, Guo L P, Ren Y Y, et al. TEM characterization of self-ion irradiation damage in Nickel-base alloy C-276 at elevated temperature. J Mater Sci Technol, 2012, 28: 1039-1045. DOI: 10.13538/j.1001-8042/nst.25.050603

[9] Jin S X, Luo F F, Ma S L, et al. Evolution of precipitate in nickel-base alloy 718 irradiated with argon ions at elevated temperature. Nucl Instrum Meth B, 2013, 307: 522-525. DOI: 10.1016/j.nimb.2012.12.105

[10] Wanderka N, Bakai A, Abromeit C, et al. Effects of 10 MeV electron irradiation at high temperature of a Ni-Mo-based Hastelloy. Ultramicroscopy, 2007, 107: 786-790. DOI: 10.1016/j.ultramic.2007.02.029

[11] Rowcliffe A F, Mansur L K, Hoelzer D T, et al. Perspectives on radiation effects in nickel-base alloys for applications in advanced reactors. J Nucl Mater, 2009, 392: 341-352. DOI: 10.1016/j.jnucmat.2009.03.023

[12] Liu W G, Han H, Ren C L, et al. The effect of Nb additive on Te-induced stress corrosion cracking in Ni alloy: a first-principles calculation. Nucl Sci Tech, 2014, 25: 050603. DOI: 10.13538/j.1001-8042/nst.25.050603

[13] Calder A F and Bacon D J. A molecular dynamics study of displacement cascades in α-iron. J Nucl Mater, 1993, 207: 25-45. DOI: 10.1016/0022-3115(93)90245-T

[14] Chen M and Hou Q. The interaction of defects in titanium: A molecular dynamics study. Nucl Sci Tech, 2010, 21: 271-274. DOI: 10.13538/j.1001-8042/nst.21.271-274

[15] Caturla M J, Soneda N, Alonso E, et al. Comparative study of radiation damage accumulation in Cu and Fe. J Nucl Mater, 2000, 276: 13-21. DOI: 10.1016/S0022-3115(99)00220-2

[16] Borodin V A and Vladimirov P V. Diffusion coefficients and thermal stability of small helium-vacancy clusters in iron. J Nucl Mater, 2007, 362: 161-166. DOI: 10.1016/j.jnucmat.2007.01.019

[17] Zhang L and Zhang Z L. Angular distribution of sputtered atoms induced by low-energy heavy ion bombardment. Nucl Sci Tech, 2004, 15: 340-343.

[18] Wang K, Xiao S F, Deng H Q, et al. An atomic study on the shock-induced plasticity and phase transition for iron-based single crystals. INT J Plasticity, 2014, 59: 180-198. DOI: 10.1016/j.ijplas.2014.03.007

[19] Ashcroft N W and Mermin N D. Solid state physics. Philadelphia, New York (USA): Thomson Learning Inc, 1976, 63-84.

[20] Yang X D, Deng H Q, Hu N W, et al. Molecular dynamics simulation of

the displacement cascades in tungsten with interstitial helium atoms. Fusion Sci Technol, 2014, 66: 112–117. DOI: 10.13182/FST13-742
[21] Yang L, Zu X T, Xiao H Y, et al. Atomistic simulation of helium-defect interaction in alpha-iron. Appl Phys Lett, 2006, 88: 091915. DOI: 10.1063/1.2178767
[22] Osetsky Yu N, Bacon D J, Serra A, et al. Stability and mobility of defect clusters and dislocation loops in metals. J Nucl Mater, 2000, 276: 65–77. DOI: 10.1016/S0022-3115(99)00170-1
[23] Wirth B D, Odette G R, Maroudas D, et al. Dislocation loop structure, energy and mobility of self-interstitial atom clusters in bcc iron. J Nucl Mater, 2000, 276: 33–40. DOI: 10.1016/S0022-3115(99)00166-X
[24] Osetsky Yu N, Bacon D J, Gao F, et al. Study of loop-loop and loop-edge dislocation interactions in bcc iron. J Nucl Mater, 2000, 283: 784–788. DOI: 10.1016/S0022-3115(00)00134-3

Note: Figure translations are in progress. See original paper for figures.