Radiation Damage in Ferritic/Martensitic Steels for Fusion Reactors: a Simulation Point of View

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Abstract. Low activation ferritic/martensitic steels are good candidates for the future fusion reactors, for, relative to austenitic steels, their lower damage accumulation and moderate swelling under irradiation by the 14 MeV neutrons produced by the fusion reaction. Irradiation of these steels, e.g. EUROFER97 which contains 8,9 % wt. Cr, 1,1 % W, 0,47 % Mn, 0,2 % V, 0,14 %Ta, 0,11 % C and Fe for the balance, is known to produce hardening, loss of ductility, shift in ductile to brittle transition temperature (DBTT) and reduction of fracture toughness and creep resistance starting at the lowest doses. The mechanisms underlying the degradation of mechanical properties are not well understood, despite numerous experimental and theoretical studies on the evolution of the microstructure under irradiation. This impedes our ability to predict and extrapolate materials' behavior at higher doses for use in the future fusion reactors. In this paper an overview on some simulations studies of irradiation induced damage accumulation and their impact on the mechanical properties is presented. In general these simulations describe so-called model alloys, as available theoretical tools cannot at this point describe real materials with their actual chemical composition. F/M steels are typically modeled by pure Fe, for it constitutes their basic structure. Helium (He), produced by transmutation by the fusion neutrons of 14 MeV, is known to impact mechanical properties, but its effect at the microstructural level is still unclear. Molecular dynamics simulations of the primary state of damage in Fe(He) and of the mobility of an edge dislocation in iron (Fe) are performed to study the effect of He, either as a gas in solid solution or in cavities.

1. Introduction

Many of the problems in the radiation-induced degradation of materials for the first wall and structural materials for the future fusion reactors require now theoretical models to span over many space and time scales in order to increase our knowledge in the field. Multiscale modeling, besides experiments, appears in this context a major tool as it allows establishing solid foundations to understanding of the relationship between irradiation damage and the induced mechanical response of the target material, starting at the atomic scale. At the basis of this modeling is a hierarchical multiscale approach that encompasses several numerical simulation methods, which include, in increasing space and time scale order, ab-initio calculations, molecular dynamics simulations and dislocation dynamics simulations. Finite element methods can then be used to predict the mechanical behavior of macroscopic specimens on the basis of the knowledge accumulated by preceding modeling. The coupling of these methods allows in principle the exploration of the outstanding problems and is part of the present modeling program of the EFDA technological program. They are used here on the Fe-He system to model the response of ferritic materials to irradiation.

Irradiation with 14 MeV neutrons leads to the formation of very short (picoseconds) and small (few tens nm) displacement cascades that give rise to vacancies, self-interstitial atoms (SIA) and spallation products such as gases (H, He) in the structure. With time these defects diffuse in the matrix and coalesce in defect clusters that will impact properties. In particular, the effect of He on the mechanical properties of low activation ferrite-based steels is an important and still unsolved issue for fusion reactor materials, as He production amounts to higher quantities than in fission reactors. Helium is known to induce certain microstructural changes in the ferritic material, such as the formation of the inconspicuous He bubbles. These changes are thought to have an impact, direct or indirect, on the mechanical behavior such as

hardening and/or embrittlement (e.g. in [1,2]). The interaction of He with irradiation induced defects such as vacancies (v), self-interstitial atoms (SIA) needs also investigation in order to complete our understanding.

Starting at the lowest scales, ab-initio type calculations are used to provide accurate information on the energetics of He atoms in Fe matrix. These data can in turn be used as reliable benchmark for molecular dynamics simulations at higher space and time scales. Molecula dynamics (MD) simulations were used in the embedded atom method to simulate the primary state of damage in Fe with He, using a combination of empirical potentials [3]. While these potentials cannot reproduce the absolute formation energies values for instance, they do reproduce qualitative trends in the behavior of He in the Fe matrix. MD simulations were used then to study the effect of He on the plasticity of Fe. It was investigated by considering that He has a direct effect on the mobility of the dislocation, either as He in solid solution of He in cavities. It was compared to the effect of a dislocation loop with a Burgers vector a_0[100]. In this paper we review the results and discuss the impact of He, firstly, on the primary state of damage and, secondly, on the plasticity as we learned from the molecular dynamics simulations.

2. Simulation Method

MD simulations are done with the code MOLDY that was originally written by Finnis [4]. It was then adopted and further developed by Diaz de la Rubia and Guinan [5] for the simulation of displacement cascades in metals. The program uses spherical many-body potentials, the embedded atom method [6] and the link cell method developed by Heyes and Smith [7], which allows accelerating the generation of the neighbour list. For all simulations the time step is 1 fs. MOLDY was used for the simulation of displacement in Fe containing He in solid solution. In order to study the interaction of a dislocation with defects we introduce in the simulation box a dislocation, which is described by a configuration of atoms whose positions are given by the anisotropic elasticity of the continuum. It is known to provide an appropriate description of the dislocation even close to the core region [8,9]. In this work the formalism of Stroh [10] is used, as it provides a compact and convenient mathematical description, which was used successfully in for instance the simulation of TEM image of dislocations [11]. In addition, we modified the MOLDY code in order to allow for deformation by shear. It can be achieved under either constant strain rate or constant stress.

The empirical potentials used for the Fe-He are those combined originally by Morishita *et al.* [3]. They combined the empirical potentials of Ackland'97 [12] for Fe-Fe, of Wilson and Johnson [13] for Fe-He and of Beck [14] for He-He. The Ackland potential is a embedded atom method type of potential, which includes a pair potential and an embedding term which accounts for the electronic density. The other two potentials, for Fe-He and He-He, are simple pair potentials. The Wilson-Johnson potential is purely repulsive and derived from Hartree-Fock-Slater calculations. The Beck potential for He-He interactions was smoothly connected with the Ziegler-Biersack-Littmark universal potential [15], which is appropriate at high energy. One might question the adequacy of these potentials to describe He in a Fe matrix. They give a formation energy of 3.25 eV for a substitutional He atom, while ab initio calculations give 4.22 eV [16]. The formation energy of the most favorable interstitial is 5.34 eV, while ab-initio gives 4.39 eV [16]. The most favorable site for an He atom is thus substitutional relative to interstitial, as indicated by both the empirical potentials and ab initio. Moreover, both types of calculations indicate that He shows self-trapping, up to 4 He atoms per vacancy [16]. However, firstly, the magnitude of the difference in formation energy by the

empirical potentials is overestimated (2.09 versus 0.17 eV for ab initio) and, secondly, the interstitial site should be tetrahedral, and not octahedral as indicated by the empirical potentials. Nonetheless, these potentials are considered to be appropriate for this work, first of all for the absence of more adequate ones. In addition, the fact that He is a noble gas indicates that no hybridization of He in Fe or charge sharing or exchange can take place. The absence of embedding term in the empirical potential with He allows reproducing this behavior.

The sample geometry for the simulation of displacement cascades in Fe-He is a cubic box 18 nm a side and containing about 0.5 million atoms. Prior to the cascade 0.1 atomic % of He is randomly distributed in interstitial sites in the body centered cubic (bcc) Fe lattice. One atom of the lattice is given the desired energy of 3 to 10 keV for the cascade and for a temperature of the lattice of 10, 300 or 523 K. Total simulation time is 25 ps, which is sufficient for a complete cool-down of the cascade. Defects are analysed using the Wigner-Seitz cell allowing for the identification of vacancies and interstitials. The crystallographic orientation of the interstitials is determined as well.

For the simulation of the deformation, the sample is 25x14x20 nm^3 in size, which corresponds to about 0.5 million atoms. The edge dislocation line is along $[11\bar{2}]$, its Burgers vector is ½ $a_0[111]$ and its glide plane is $(1\bar{1}0)$. The defects are a 2 nm cavity or a 2 nm $a_0[100]$ loop. Simulations are performed at 10 K for the clarity of the atomic configurations during the simulation. Simulations were done under constant strain rate and the stress response was recorded. The stress response in the case of the dislocation interacting with a defect gives thus the release or escape stress at the moment when the dislocation escapes from the defect. The release stress is equivalent to the obstacle strength.

3. Results

The main results of the displacement cascades in Fe and Fe containing 0.1 % He in solid solution simulated by MD are summarized here. The detail should be published elsewhere [17]. It appears that the produced vacancies remain in a central region of the displacement cascade and the presence of He does not influence vacancy and SIA production. While 0.1 % He in interstitial and substitutional sites does not seem to affect the collisional stage of the cascade, it does determine final damage. The defect production efficiency in Fe-0.1 % He in interstitial sites is slightly higher than that in Fe-0.1 % He in substitutional sites and in pure Fe. Most interstitials are Fe-Fe and Fe-He dumbbells, in the <110> and <100> crystallographic direction, respectively. The number and size of interstitial clusters is larger with interstitial He than with substitutional He and than with pure Fe. These latter results, in pure Fe, are in agreement with results of other authors [18,19,20]. The number and size of interstitial clusters increase with the PKA energy and temperature in the range 10K to 523K. The striking result of this study is that the interstitial clusters in small and medium size contain a large percentage of He atoms, which stabilize them. The fraction of He to Fe in interstitial clusters is mostly 50 %. It appears that during the cascade evolution all displaced Fe and He atoms combine with each other to form either Fe-He or Fe-Fe interstitial pairs as well as interstitial clusters. Only a small number of large clusters consisting of a large number of Fe interstitials and only a few He atoms are produced, the majority being clusters with small and medium sizes containing an equal amount of Fe and He.

Our results clearly indicate a strong synergy between the He atoms and the point defects. In effect, He atoms join with SIA to form clusters, and stabilize them. It should be noted that there is no vacancy near these clusters or combined with He. These features would drastically

reduce the mobility of interstitial clusters, which in turn would allow the growth of vacancy clusters by impeding annihilation.

The MD simulation of the plasticity of Fe containing an edge dislocation interacting with defects shows the following. The detail of this study will appear elsewhere [21]. Helium was considered to have a direct effect on the mobility of the dislocation. It appeared though that the presence of substitutional He in the sample does not change significantly the yield stress or the flow stress, even at the highest He content considered of 1.0 at. %. Helium can segregate and form bubbles. To study their effect we considered firstly a void and secondly a bubble of the same size of 2 nm with varying content in He. It appears that the void is a stronger obstacle than the He bubble at low He contents. The void gives rise to an obstacle strength of 590 MPa. This is comparable to what is found by other authors [22]. As the cavity is filled with He its strength decreases to a minimum of 350 MPa when it contains 2 He atoms per vacancy. The release strain is also reduced, to about half the value needed for the void. As the He content increases the bubble strength and release strain increase. At 5 He atoms per vacancy, the bubble presents a resistance of 800 MPa to the impinging edge dislocation.

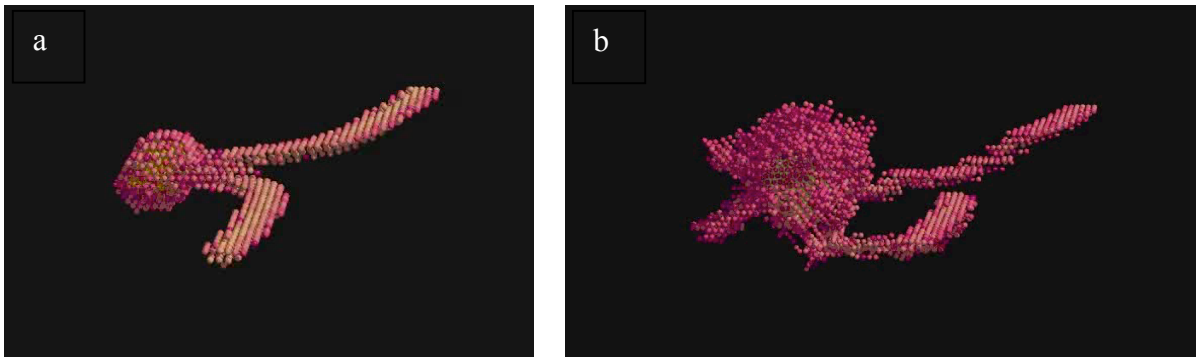

Fig. 1. Perspective view of the MD simulation of the moving dislocation in Fe pinned and bowing at (a) a 2 nm void, and at (b) a 2 nm He bubble containing 5 He atoms per vacancy. Dislocation segments emission from the bubble (b) due to high internal pressure is visible.

Figure 1 illustrates the case of the edge dislocation impinging on a 2 nm void (Fig. 1a) and on a 2 nm He bubble (Fig. 1b) containing 5 He atoms per vacancy. The dislocation is pinned and bows out under the applied strain. Clearly in the case of the He bubble we observe dislocation segments that were punched out from the surface of the bubble due to the pressurized He gas. This so-called loop punching explains the high obstacle strength exhibited by the He bubble.

In the literature there is experimental evidence of gas bubble induced loop punching, such as for H bubbles in Cu [23] and He bubbles in Al-0.4Li [24]. Ab-initio calculations do indicate that such an effect might be also effective in Fe, as they show that up to 4 He atoms strongly bind with a single vacancy [16]. This so-called self-trapping of He [25] was however to our knowledge never observed in ferritic materials.

The void constitutes a hard obstacle to the moving dislocation. Conversely, He in solid solution does not have any significant direct effect on mobility, and when it is added in the void to form a He bubble, the void looses its obstacle's strength. However, when He content exceeds a critical value, which is highly improbable in reality, dislocation segments are emitted around the bubble, which strengthen it (Fig. 1). To summarize, results indicate that He, either in the form of He bubbles or in solid solution, does not have a direct effect on the

mechanical properties. When considering the results from the displacement cascade it appears that if He has an effect on the mechanical properties it may be an indirect one, by promoting the formation of other types of clusters.

4. Conclusions

Molecular dynamics simulations of the primary state of damage in Fe(He) show that the presence of He in interstitial sites promote the formation of self interstitial atom clusters. Indeed, their number and size is drastically increased when compared to the case of pure Fe or Fe with He in substitutional sites. Moreover, He tends to combine with SIAs and the fraction of He to Fe in interstitial clusters is mostly 50 %. This feature may drastically reduce the mobility of interstitial clusters in Fe-0.1 % He interstitial state compared to pure Fe, with significant impact on the subsequent evolution of radiation induced defects in the alloy. In effect, vacancy clusters would grow more easily due to the reduced annihilation rate. This in turn will have a significant impact on mechanical properties, as we know from MD simulations that voids are strong obstacles to moving dislocations. The MD simulations show that He in solid solution doesn't have a significant effect on the yield stress before it reaches 1.0 at. % He. The 2 nm void presents an obstacle strength of 590 MPa to the edge dislocation. A 2 nm He bubble is a weaker obstacle than the 2 nm void when the He content is low, at 1 and 2 He atoms per vacancy. Beyond 2 He atoms per vacancy, a content at which the bubble is the weakest, the resistance of the He bubble increases with increasing He content. At 5 He atoms per vacancy, the He bubble becomes a much stronger obstacle, which is due to significant loop punching.

It appears thus that in this scenario, the produced He, forming and stabilizing SIA-He clusters has at most an indirect effect on hardening and/or embrittlement. The main effect of He would thus reside on the migration of point defect, interstitials and vacancies, which in turn would modify the density and size of voids, which are strong obstacles. Another effect that could be induced by He and that would be worth studying by simulations is intergranular fracture, which promotes embrittlement.

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4. References

[1] KLUEH, R., HARRIES, D., 'High-Chromium Ferritic and Martensitic Steels for Nuclear Applications', American Society for Testing and Materials, 2001.

[2] YAMAMOTO, T., et al., "On the effects of irradiation and helium on the yield stress changes and hardening and non-hardening embrittlement of ~8Cr tempered martensitic steels: Compilation and analysis of existing data", J. Nucl. Mater. **356** (2006) 27.

[3] MORISHITA, K., et al., "Thermal stability of helium-vacancy clusters in iron", Nucl. Instr. and Meth. in Phys. Res. B **202** (2003) 76.

[4] FINNIS, M.W., MOLDY6 – A Molecular Dynamics Program for Simulation of Pure Metals, Harwell Report AERE-R-13182 (1989).
[5] DIAZ DE LA RUBIA, T., GUINAN, M.W., "Progress in the development of a molecular-dynamics code for high-energy cascade studies", J. Nucl. Mater. **174** (1990) 151.
[6] DAW, M. S., BASKES, M.I., "Embedded-atom method: derivation and application to impurities, surfaces, and other defects in metals", Phys. Rev. B **29** (1984) 6443.
[7] HEYES, D.M., in: Information Quarterly for Computer Simulation of Condensed Phases, No 26, Science and Engineering Research Council, Daresbury Laboratory, Daresbury, Warrington WA4AD, England (1987) p. 68.
[8] MILLS, M.J., STADELMANN, P. , "A study of the structure of lomer and 60-degrees dislocations in aluminium using high resolution transmission electron microscopy", Philos. Mag. A **60** (1989) 355.
[9] BALK, T.J., HEMKER, K.J., "High resolution transmission electron microscopy of dislocation core dissociations in gold and iridium", Philos. Mag. A **81** (2001) 1507.
[10] STROH, A.N., "Dislocations and cracks in anisotropic elasticity", Philos. Mag. (1959) 625.
[11] SCHAEUBLIN, R., STADELMAN, P., "A method for simulating electron microscope dislocation images", Mater. Sci. Eng. A **164** (1993) 373.
[12] ACKLAND, G.J., et al., "Computer simulation of point defect properties in dilute Fe-Cu alloy using a many-body interatomic potential", Philos. Mag. A **75** (1997) 713.
[13] WILSON, W.D., JOHNSON, R.D., Rare gases in metals, in: P.C. Gehlen, J.R. Beeler Jr., R.I. Jaffee (Eds.), Interatomic Potentials and Simulation of Lattice Defects, Plenum, 1972, p. 375.
[14] BECK, D.E., "A new interatomic potential function for helium", Mol. Phys. **14** (4) (1968) 311.
[15] BIERSACK, J.P., ZIEGLER, J.F., "Refined universal potentials in atomic-collisions", Nucl. Instrum. Meth. **194** (1982) 93.
[16] FU, C.C., WILLAIME, F., "Ab initio study of helium in alpha-Fe: Dissolution, migration, and clustering with vacancies", Phys. Rev. B **72** (2005) 064117.
[17] YU, J., et al., "Synergistic effect of PKA and He in Fe-0.1 % He matrix", submitted for publication to J. Nucl . Mater.
[18] MALERBA, L., et al., "Molecular dynamics simulation of displacement cascades in Fe-Cr alloys", J. Nucl. Mater. **329-333** (2004) 1156.
[19] STOLLER, R.E., "The role of cascade energy and temperature in primary defect formation in iron", J. Nucl. Mater. **276** (2000) 22.
[20] BECQUART, C., et al., "Influence of the interatomic potentials on molecular dynamics simulations of displacement cascades", J. Nucl. Mater. **280** (2000) 73.
[21] SCHAEUBLIN, R., CHIU, Y.L., "Effect of helium on irradiation-induced hardening of iron: a simulation point of view", accepted for publication in J. Nucl. Mater.
[22] OSETSKY, Y.N., BACON, D.J., "Comparison of void strengthening in fcc and bcc metals: Large-scale atomic-level modeling", Mater. Sci. Eng. A **400-401** (2005) 374.
[23] WAMPLER, W.R., et al., "Precipitation and trapping of hydrogen in copper", Philos. Mag. **34** (1) (1976) 129.
[24] SHIRAISHI, K., et al., "Radiation-generated prismatic loops around gas-bubbles in alumnium-lithium alloy", Radiat. Eff. **21** (3) (1974) 161.
[25] WISLON, W.D., et al., "Self-trapping of helium in metals", Phys. Rev. B **24** (10) (1981) 5616.