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Effect of Cr content on the nanostructural evolution of irradiated ferritic/martensitic alloys: An object kinetic Monte Carlo model

M. Chiapetto [a, b, *], L. Malerba [a], C.S. Becquart [b]

[a] SCK•CEN, Nuclear Materials Science Institute, Boeretang 200, B-2400 Mol, Belgium
[b] Unité Matériaux Et Transformations (UMET), UMR 8207, Université de Lille 1, ENSCL, F-59600 Villeneuve d'Ascq Cedex, France

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ABSTRACT

Self-interstitial cluster diffusivity in Fe–Cr alloys, model materials for high-Cr ferritic/martensitic steels, is known to be reduced in a non-monotonic way as a function of Cr concentration: it first decreases, then increases. This non-monotonic behaviour is caused by a relatively long-ranged attractive interaction between Cr atoms and crowdions and correlates well with the experimentally observed swelling in these alloys under neutron irradiation, also seen to first decrease and then increase with increasing Cr content, under comparable irradiation conditions. Moreover, recent studies reveal that C atoms dispersed in the Fe matrix form under irradiation complexes with vacancies which, in turn, act as trap for one-dimensionally migrating self-interstitial clusters. The mobility of one-dimensional migrating clusters is considered key to determine swelling susceptibility. However, no model has ever been built that quantitatively describes the dependence of swelling on Cr content, allowing for the presence of C in the matrix. In this work we developed physically-based sets of parameters for object kinetic Monte Carlo (OKMC) simulations intended to study the nanostructure evolution under irradiation in Fe–Cr–C alloys. The nanostructural evolution in Fe–C and in four Fe–Cr–C alloys (containing 2.5, 5, 9 and 12 wt.% Cr) neutron irradiated up to ~0.6 dpa at 563 K was simulated according to the model and reference experiments were reproduced. Our model shows that the SIA cluster reduced mobility has a major influence on the nanostructural evolution: it increases the number of vacancy-SIA recombinations and thus leads to the suppression of voids formation. This provides a clear framework to interpret the non-monotonic dependence of swelling in Fe–Cr alloys versus Cr content. Our model also suggests that the amount of C in the matrix has an equally important role: high amounts of it may counteract the beneficial effect that Cr has in reducing swelling.

1. Introduction

High-Cr ferritic/martensitic (F/M) steels are candidate structural materials in the breeding blanket of future fusion reactors [1], as well as for fuel cladding and other core components in GenIV reactors [2,3]. The Cr content in these steels is known to have a strong effect on radiation defect stability and, in general, on the properties of the material. For this reason, Fe–Cr alloys are often employed as model materials in irradiation experiments and subsequent characterization [4–20]. It is known in particular from these studies that the addition of Cr to Fe reduces radiation-induced swelling, typically by an order of magnitude [4,6–9]. Specifically, swelling drastically decreases with the addition of small quantities of Cr (few wt.%), remains low between ~1 and 10 %Cr and then rises again, although the actual behaviour and thresholds depend strongly on the irradiation conditions. For example swelling local maxima are observed at very high irradiation dose around 9 %Cr [4–9]. Cr content is therefore a key parameter, the effect of which needs to be fully understood in order to guarantee the best swelling resistance, together with minimum embrittlement, and cope with the high energy neutron dose in the harsh environmental conditions expected in future nuclear systems. With this aim, international efforts are being made to develop physically-based models capable of describing the behaviour of Fe–Cr alloys under irradiation as a function of irradiation parameters (fluence, temperature, …) and Cr content [21–30].

* Corresponding author. SCK•CEN, Nuclear Materials Science Institute, Boeretang 200, B-2400 Mol, Belgium.
E-mail address: mchiapet@sckcen.be (M. Chiapetto).

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Both experimental [11–14] and theoretical [26–30] studies, the latter making use of atomistic computer simulation techniques, suggest that one of the main effects of Cr on the nanostructural evolution under irradiation of Fe–Cr alloys is the reduction of the mobility of self-interstitial clusters (dislocation loops) versus temperature, especially for 1/2⟨111⟩ loops [16]. This mobility reduction, physically due to the attractive interaction between the Cr atoms dispersed in the solid solution and the crowdions that form the clusters, has been theoretically shown to exhibit the same non-monotonic dependence on Cr content as empirically observed in void swelling suppression (cfr [4,6–8]. and [27–30]). However, up to now the connection between slowing down/stabilization of interstitials clusters due to Cr in Fe and swelling suppression in Fe–Cr alloys as compared to Fe has been argued, but not demonstrated. Moreover, the effect of Cr in the past has been assessed while completely disregarding the fact that C is also contained not only in steels but also, unavoidably, in any iron-based model alloys. C modifies the phase diagram of the Fe–Cr system, so its presence combined with the actual Cr content will determine microstructural changes (eg ferrite versus martensite, carbide formation, …) which will impact the actual content of C in the matrix, quite obviously with consequences on the nanostructural evolution under irradiation. Finally, the nanostructural evolution is obviously also dependent on the presence and amount of sinks related to the initial microstructure, namely dislocations and grain boundaries. Here we develop a model capable of simulating the nanostructural evolution in Fe–Cr–C alloys with different Cr content, neutron-irradiated at ~290 °C up to doses of the order of ~1 dpa, taking into account, under appropriate approximations, also the effect of the presence of C and its concentration in the matrix, while introducing sensible descriptions of the sinks present in the material. This was possible by using as starting point a nanostructure evolution model recently developed for Fe–C alloys [31,32]. More in detail, we investigate Fe–C alloys with four Cr concentrations (2.5, 5, 9 and 12 wt.%Cr, nominal compositions), by simulating their neutron irradiation up to ~0.6 dpa at 563 K. The results are compared with experimental data obtained on real alloys, irradiated under the mentioned conditions, stemming from the thorough microstructural examination these alloys have undergone, that was completed over the past years [9,10,15–19,33]. As there is no single experimental technique capable of giving a complete description of the irradiation-induced microstructure of steels and Fe-based model alloys, a combination of techniques sensitive to defects at the nm-scale, such as transmission electron microscopy (TEM) [10,16], atom probe tomography (APT) [19], small angle neutron scattering (SANS) [17,18] and positron annihilation spectroscopy (PAS) [9], was used in order to thoroughly characterize the Fe–Cr–C alloys in terms of defect densities and average sizes of both vacancy and SIA cluster populations at different doses. With our model, by comparison with this wide range of experimental results, we show that, indeed, the effect of Cr on interstitial cluster properties can explain the nanostructure evolution observed in these alloys under irradiation; we also reveal the importance of the actual amount of C in the matrix to determine the evolution of the populations of loops and voids.

The paper is organized as follows: the simulation method is presented in Section 2, while Section 3 outlines the main aspects of the adopted parameterization, together with the influence of Cr content on the mobility of the interstitial cluster population. In Section 4 we show the simulation results, in terms of evolution of both vacancy and interstitial defect populations: they are compared with the corresponding experimental observations. Finally, Section 5 contains a discussion of our results together with an overview on the main limitations of the current OKMC model. In Section 6 our conclusions are summarized.

2. Computational method

For all our simulations we used the object kinetic Monte Carlo (OKMC) code LAKIMOCA, thoroughly described in Ref. [34]. The approach we adopt is explained in detail in Refs. [31,32]. For convenience, we highlight here the fundamental ideas. The OKMC is a stochastic method used to describe the evolution of defects and their clusters in materials subjected to irradiation and/or annealing, disregarding the detail of processes directly involving atoms and focusing instead only on the properties of defects, treated as objects. Irradiation produces point-defects, namely vacancies (V) and self-interstitial atoms (SIA), which may form clusters. These clusters accumulate in growing number densities and size and may also form complexes with specific solute atoms, if present. The objects we consider are therefore V and SIA clusters or any other nanostructural feature, like carbon (C) atoms or carbon-vacancy (C_mV_n) complexes, located in a simulation volume in which the object coordinates are known and tracked. It is, however, important to note that, in the case of the Fe–Cr–C alloys addressed here, Cr atoms are not treated as objects, because this would lead to an unnecessarily large number of elements in the system, that would slow down enormously the simulation. Instead, the effect of Cr is introduced assuming that it changes the properties of the objects, i.e. applying a “grey alloy” approach.

Each of the objects introduced in the system can undergo events such as migration, recombination, or clustering of defects, which take place in the simulation volume (or box) according to predefined probabilities. Every object has an associated reaction volume, that is generally a sphere, with the exception of large dislocation loops (>150 SIA), which are represented by toroids. When the reaction volumes of two objects overlap, a predefined reaction, like clustering between two vacancy clusters, or annihilation between a vacancy and an SIA, takes place. The events in the OKMC simulation determine the dynamics of the system. The probability for the objects to perform an event are given in terms of Arrhenius frequencies for thermally activated processes, according to transition state theory:

$$\Gamma_i = \nu_i exp\left(\frac{-A_i}{k_B T}\right) \tag{1}$$

Here ν_i is the attempt frequency (alias the prefactor) of the event i; A_i is the corresponding activation energy, which must embody both the thermodynamics and the kinetics of the system being studied; k_B is the Boltzmann's constant and T is the irradiation temperature expressed in K. Given that the activation energy for migration or emission depends on type and size of the objects, a very large number of parameters is needed for a standard simulation. For every simulation step, among all the possible events, one and only one is chosen, based on the corresponding probabilities in the parameterization and according to the stochastic Monte Carlo algorithm [35]. Time elapses according to the residence time algorithm [36,37]:

$$\Delta t \propto \frac{1}{\sum_1^N \Gamma_i} \tag{2}$$

where the time increase is obtained as inverse of the sum of all the frequencies Γ_i associated with each of the N possible events.

A special kind of objects employed in our model are traps and sinks. These are immobile spherical objects that can, respectively, trap mobile defects (vacancy and SIA clusters) with a certain binding energy, E_t^δ, that depends on the size and type of the trapped object, or remove them from the system. In the work presented here, traps are used to simulate the effect of carbon or carbon-