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Materials subjected to fast neutron irradiation/Matériaux soumis à irradiation par neutrons rapides

First principles calculations in iron: structure and mobility of defect clusters and defect complexes for kinetic modelling

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Abstract

Predictive simulations of the defect population evolution in materials under or after irradiation can be performed in a multi-scale approach, where the atomistic properties of defects are determined by electronic structure calculations based on the Density Functional Theory and used as input for kinetic simulations covering macroscopic time and length scales. Recent advances obtained in iron are presented. The determination of the 3D migration of self-interstitial atoms instead of a fast one-dimensional glide induced an overall revision of the widely accepted picture of radiation damage predicted by previously existing empirical potentials. A coupled ab initio and mesoscopic kinetic Monte Carlo simulation provided strong evidence to clarify controversial interpretations of electrical resistivity recovery experiments concerning the mobility of vacancies, self-interstitial atoms, and their clusters. The results on the dissolution and migration properties of helium in α-Fe were used to parameterize Rate Theory models and new inter-atomic potentials, which improved the understanding of fusion reactor materials behavior. Finally, the effects of carbon, present in all steels as the principal hardening element, are also shown. ***To cite this article: C.C. Fu, F. Willaime, C. R. Physique 9 (2008).***

Résumé

Calculs ab initio dans le fer : structure et mobilité des agrégats de défauts pour modélisation cinétique. Les simulations prédictives de l'évolution de la population de défauts dans un matériau sous ou après irradiation peuvent être effectuées dans une approche multi-échelle, où les propriétés à l'échelle atomique sont déterminées par des calculs de structure électronique basés sur la Théorie de la Fonctionnelle de la Densité puis utilisées comme données d'entrée dans des simulations de cinétique couvrant des échelles de temps et d'espace macroscopiques. Nous présentons les avancées récentes obtenues dans ce domaine dans le fer. Suite à la mise en évidence d'un mécanisme de migration tri-dimensionnel des auto-interstitiels au lieu d'un glissement rapide unidimensionnel, le scénario de l'évolution des défauts d'irradiation basé sur les prédictions venant des potentiels empiriques a été complètement révisé. Un couplage entre calculs ab initio et simulations de Monte Carlo cinétique a permis de clarifier l'interprétation controversée des expériences de recuit de résistivité, en ce qui concerne la mobilité des lacunes et interstitiels et de leurs amas. Les résultats sur les propriétés de mise en solution et de migration de l'hélium dans le fer α, ont été utilisés comme données d'entrée de modèles de cinétique chimique ainsi que pour ajuster des nouveaux potentiels empiriques. Les études associées contribuent à une meilleure compréhension du devenir de l'hélium dans les matériaux des centrales nucléaires à fusion. Enfin, nous montrons l'effet que le carbone, présent dans tous les aciers comme principal élément durcissant, peut avoir sur les phénomènes étudiés. ***Pour citer cet article : C.C. Fu, F. Willaime, C. R. Physique 9 (2008).***

Keywords: First principles; Iron; Helium; Carbon; Radiation damage; Defects

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Mots-clés : Calculs ab initio ; Fer ; Hélium ; Carbone ; Dégât par rayonnement ; Défauts

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1. Introduction

Ferritic steels play a central role in nuclear technology, in particular as structural materials for fission and future fusion nuclear reactors. Although their mechanical properties have been extensively investigated, little is known about the structural, electronic and magnetic properties at the origin of their macroscopic behavior. First principles calculations within the Density Functional Theory (DFT) provide such information at the atomic scale, which is not directly accessible through experiments. However, the application of first principles studies in this field is rather new compared with other Solid State Physics and Material Science disciplines. The objective of this article is to report key results of recent DFT studies which either contributed directly to reconcile theory and experiments, or served as input data for kinetic models, or to parameterize empirical interatomic potentials.

This paper is a review of the most recent results for the particular case of intrinsic defects and He in α-Fe. It does however, not pretend to be a complete review of all existing ab initio data, but only a few representative examples revealing the predictive power and utility of first principles methods for the study of radiation damage. Except where otherwise mentioned, all the results reported here have been obtained using the DFT-SIESTA code (Spanish Initiative for Electronic Simulations with Thousands of Atoms: http://www.uam.es/siesta) [1], generalized gradient approximation (GGA), non-local norm-conserving pseudopotentials, and strictly localized pseudo-atomic basis functions. The errors in energy differences (e.g. migration or binding energies) related to the convergence of k-point grids and real space grids for charge density representation are estimated to be less than 0.05 eV in the present approach.

In the next section of the paper we review the main energetic and kinetic properties of vacancies self-interstitial atoms according to ab initio results. In the third section, the results on the dissolution, migration of He atoms, and their interaction with intrinsic defects are reported. The possible effects of carbon on these results are discussed in the fourth section.

2. Intrinsic defects and their clusters

The microstructural changes of nuclear materials are mainly governed by the properties of defects produced by irradiation. The information on the mobility, recombination, clustering or dissociation of vacancies (V), self-interstitial atoms (SIAs) and their clusters can, however, be obtained only indirectly by experiments such as macroscopic electrical resistivity recovery measurements [2], which provide extremely rich information but which is difficult to interpret. By combining ab initio and kinetic Monte Carlo methods, we successfully reproduced the abrupt resistivity changes—the resistivity recovery stages—observed upon annealing at increasing temperatures after electron irradiation in α-iron [3].

Ab initio results obtained for SIA (i.e. mono-interstitial) stabilities and migration energies in Body Centered Cubic (bcc) iron are indeed in good agreement with the above mentioned experiments [4,5]. Studies prior to 2003 based on empirical potentials predict that the $\langle 110\rangle$ and $\langle 111\rangle$ dumbbell configurations of SIAs are very close in energy, and that SIAs migrate via very fast one-dimensional motion, combined with occasional thermally activated reorientations [6–8]. This widely accepted picture has been questioned by ab initio calculations showing that the energy difference between the ground state $\langle 110\rangle$ and the $\langle 111\rangle$ dumbbells is much larger than expected, around 0.7 eV [4]. This large value makes a migration via the $\langle 111\rangle$ dumbbell incompatible with the experimental value of the SIA migration energy of 0.3 eV [2]; we have therefore searched and identified by means of DFT that the most favorable migration mechanism corresponds to the nearest-neighbor translation–rotation jump (Fig. 1), with a migration energy, of 0.34 eV in excellent agreement with the experimental value of 0.30 ± 0.03 eV [9]. This mechanism, originally suggested in Ref. [10], implies three-dimensional migration of SIAs in agreement with experimental evidence. The ab initio SIA formation energies have then been used by Mendelev et al. to parameterize a new EAM potential in 2003 which gives an improved description of self-interstitial atoms and clusters [11].

We have also performed studies on the stability and migration properties of clusters containing up to either three SIAs or five vacancies by searching the most stable configurations, and exploring several possible migration mechanisms. Small interstitial-type clusters made of parallel $\langle 110\rangle$ single dumbbells are found to have low formation energies, with e.g. for the quadri-interstitial a structure where the $\langle 110\rangle$ dumbbells form a compact rhombus in the

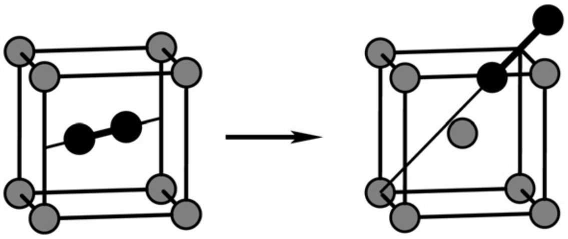

Fig. 1. Schematic view of the translation–rotation mechanisms of a ⟨110⟩ dumbbell.

Fig. 1. Représentation schématique du mécanisme de migration par translation–rotation du dumbbell ⟨110⟩.

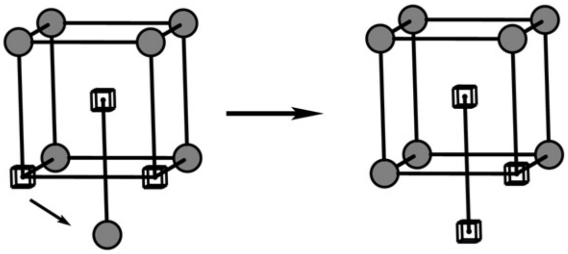

Fig. 2. Schematic view of the low energy migration mechanism of a tri-vacancy cluster. The empty cubes represent the vacancies.

Fig. 2. Représentation schématique du mécanisme de migration de plus basse énergie de la tri-lacune. Les cubes représentent les lacunes.

(110) plane. The binding energies defined as the energy gain due to the last defect joining the cluster are 0.80, 0.82 and 1.64 eV for the di-, tri-, and quadri-interstitial, respectively. Di- and tri-interstitials have almost the same migration energies (0.42–0.43 eV) by performing the same translation–rotation jump as in the mono-interstitial case. Clusters containing more than four SIAs are found to have lower energies when they are made of ⟨111⟩ dumbbells rather than of parallel ⟨110⟩ ones [12]. Both experimental evidences and empirical potential based Molecular Dynamics studies have shown that ⟨111⟩ clusters perform mainly one-dimensional migration [6–8,13].

Vacancy clusters are less stable than the interstitial ones. Small vacancy clusters (V_n) are found to have the well-known ground state configurations [14,15], the corresponding binding energies for V_{2-5} are 0.30, 0.37, 0.62 and 0.73 eV respectively. They migrate by successive nearest-neighbor mono-vacancy jumps. The migration energies are size dependent: 0.67, 0.62, 0.35 and 0.48 eV for $n = 1, 2, 3$ and 4 respectively. We emphasize that unexpected low values are obtained for V_3 and V_4, unlike in earlier studies. Larger vacancy clusters were believed to be also mobile, but with a mobility decreasing with size: e.g. the migration energy of V_5 was estimated to be 0.83 eV.

These ab initio binding and migration energies, together with some additional assumptions described in detail in Ref. [3], were used as input data for an event-based kinetic Monte Carlo calculation to simulate resistivity recovery experiments after electron irradiation. As a result, all the recovery stages were successfully reproduced and interpreted. New features in the mechanisms responsible for these stages were revealed. In particular, we provided strong evidence that the controversial stage (III) is due to the migration of vacancy-type clusters. We have shown that V_{2-4} clusters have migration energies lower than that of a mono-vacancy: they are therefore immediately mobile when formed as a result of vacancy migration at stage (III). These clusters, in particular V_2, were suggested to be the anisotropic defects detected by Magnetic After Effect (MAE) experiments at stage (III). Concerning interstitial-type clusters, we confirmed the compatibility of the translation–rotation mechanism of ⟨110⟩ dumbbell predicted by DFT and the experimental results, we have also shown that both di- and tri-interstitials contribute to stage (II) even at low irradiation doses.

3. Behavior of He atoms in iron and their interaction with self-defects

In addition to the intrinsic defects, large amounts of He and H are also produced by nuclear transmutation under high energy neutron irradiation. As a first step to understand the behavior of structural materials for future fusion reactors, the energetics and kinetics of He atoms and their interaction with point defects must be understood. Some

Table 1
Solution properties of He in α-Fe. Comparison between DFT-SIESTA and empirical potential (EP) results: Ackland 97 Finnis Sinclair potential for Fe–Fe, and either the Wilson or the new Juslin–Nordlund potential for the Fe–He interaction. All the solution energies ($E(\mathrm{He}_{\mathrm{sub}})$, $E(\mathrm{He}_{\mathrm{tetra}})$ and $E(\mathrm{He}_{\mathrm{octa}})$) are in electronvolts

Property	DFT-SIESTA	EP (Wilson)	EP (Juslin)
$E(\mathrm{He}_{\mathrm{sub}})$	4.22	3.25	4.04
Preferential int. site	tetrahedral	octahedral	tetrahedral
$E(\mathrm{He}_{\mathrm{tetra}})$–$E(\mathrm{He}_{\mathrm{sub}})$	0.17	2.09	0.31
$E(\mathrm{He}_{\mathrm{tocta}})$–$E(\mathrm{He}_{\mathrm{tetra}})$	0.18	−0.09	0.11

ab initio studies have also been made for this purpose [16–18]. We have performed calculations on the stability of He in substitutional and interstitial sites, denoted $\mathrm{He}_{\mathrm{sub}}$ and $\mathrm{He}_{\mathrm{int}}$ hereafter, on the migration energies of He, and on the interaction of He with vacancies and SIAs in α-Fe. Our results on the relative stabilities of substitutional and interstitial sites are in good agreement with VASP calculations based on plane-wave basis sets from Seletskaia et al. [16]. Both DFT studies predict that the most favorable solution site for helium is the substitutional site, followed by the tetrahedral and then the octahedral sites. The energy difference between the substitutional and the tetrahedral site is small, i.e. around 0.2 eV. These results are significantly different from those from previous empirical potential ones using the Wilson's Fe–He potential [19]. According to such results, the octahedral site is more stable than the tetrahedral one, and more importantly a much larger difference between the substitutional and the preferential interstitial site has been predicted. These discrepancies have direct effects on the diffusion properties of substitutional He in iron [17]. Based on ab initio results, a new pairwise Fe–He potential has been developed recently by Juslin and Nordlund [20] in order to fix this problem. A summary of the solution properties is given in Table 1.

We want to point out that, on one hand, the ab initio energy difference between the tetrahedral and the octahedral sites is indeed a robust result, in the sense that it is practically independent of all the approximation made: i.e. the exchange–correlation functional, the pseudopotential, the treatment of semi-core states and the basis sets. Also, it is not a specificity of Fe but a common property of all the bcc transition metals [21]; it is therefore not related to magnetism as previously suggested in Ref. [16]. On the other hand, the DFT energy difference between the substitutional and the tetrahedral value was shown to be more sensitive to the exchange-correlation functional and the pseudopotential used [21], its error bar may be as large as 0.15 eV.

In order to understand the long range diffusion of He, the elementary mechanisms of He migration must be known. The possible mechanisms—the vacancy, dissociative, replacement (kick-out), and the exchange mechanism—have been proposed for decades and described by Trinkaus et al. in a review paper [22]. They are also detailed in the article by Caturla et al. in this issue [23].

We focus here on the vacancy and dissociative mechanisms for a substitutional He. The first mechanism requires another incoming vacancy. We obtained a V to HeV binding energy of 0.78 eV with two vacancies being first neighbors and the He atom located midway between them. Two two-step jump mechanisms were proposed for the migration of the HeV_2 complex [17], the lowest energy mechanism consists in a nearest neighbor jump of one of the two vacancies transforming the nearest neighbor configuration into a third neighbor one. In this metastable configuration He occupies a substitutional site. Then, by a similar but reverse jump, a nearest neighbor configuration is recovered. The corresponding migration energy is 1.08 eV. This energy barrier is lower than the lower bound value of the V from HeV_2 dissociation energy (1.45 eV), estimated from the sum of the V to He–V binding energy and the V migration energy (0.67 eV). Therefore the migration of substitutional He by vacancy mechanism is expected to be governed by HeV_2 complex migration.

Substitutional He can also migrate via a dissociative mechanism i.e. when a substitutional He dissociates from its vacancy to migrate through interstitial sites until trapping at another vacancy. The corresponding process can be described by the following reaction:

$$\mathrm{He}_{\mathrm{int}} + \mathrm{V} \leftrightarrow \mathrm{He}_{\mathrm{sub}}$$

The two important magnitudes for the dissociation mechanism are therefore the interstitial He migration energy, and the energy difference between the $\mathrm{He}_{\mathrm{sub}}$ and the $\mathrm{He}_{\mathrm{int}}$ (Table 1). The former value is given by the $\mathrm{He}_{\mathrm{tetra}}$ migration energy. A tetrahedral He can migrate between two equivalent sites without passing through an octahedral one. We find a very low energy barrier, namely 0.06 eV, similar to the value found with empirical potential of 0.08 eV [17,19],

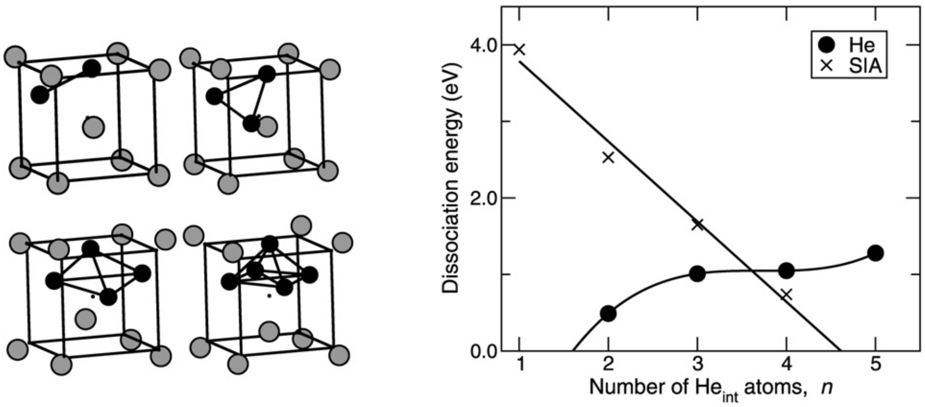

Fig. 3. Left: schematic view of interstitial He$_n$ clusters with $n = 2, 3, 4$ and 5, where the atoms are in their optimized positions. Right: dissociation energies of a He$_{\text{int}}$ or a SIA from He$_n$ clusters.

Fig. 3. A gauche : représentation schématique des amas d'hélium intersitiel, He$_n$, avec $n = 2, 3, 4$ et 5. Les atomes sont représentés à leur position relaxée. A droite : énergie de dissociation d'un atome d'hélium interstitiel ou d'un auto-interstitiel à partir d'un amas He$_n$.

although the latter value corresponds to a jump between two nearest neighbor octahedral sites. Concerning the latter value, DFT predicted a low energy difference which makes it possible for the dissociation mechanism to occur.

The dissociation mechanism will obviously dominate over the vacancy mechanism when the concentration of free migrating vacancies is low, e.g. under thermal equilibrium condition, or under irradiation when most of the vacancies are either trapped by impurities (e.g. C, N) or eliminated at sinks such as surfaces or grain boundaries. On the other hand, the dissociative mechanism will dominate when there is super-saturation of free vacancies. More details and expressions for the effective migration energies corresponding to limiting cases are reported in Ref. [17].

The stability of small He$_n$V$_m$ clusters was also investigated for n and $m = 0$ to 4. The search for the ground state configurations was performed by relaxing for each cluster size a set of candidate compact arrangements of vacancies filled with He. The binding energies of a vacancy to the clusters can be obtained with the following expression:

$$E^{\mathrm{B}}(V) = E\big(\big[N-(m-1)\big]\mathrm{Fe}, n\mathrm{He}\big) + E\big((N-1)\mathrm{Fe}\big) - E\big((N-m)\mathrm{Fe}, n\mathrm{He}\big) - E(N\mathrm{Fe})$$

where $E((N-m)\mathrm{Fe}, n\mathrm{He})$ is the energy of the system with $(N-m)$Fe atoms and a He$_n$V$_m$ cluster. The binding energy of an interstitial tetrahedral helium atom with a He$_{n-1}$V$_m$ can be defined in a similar way as:

$$E^{\mathrm{B}}(\mathrm{He}) = E\big((N-m)\mathrm{Fe}, (n-1)\mathrm{He}\big) + E(N\mathrm{Fe}, \mathrm{He}) - E\big((N-m)\mathrm{Fe}, n\mathrm{He}\big)$$

They correspond respectively to the reactions:

$$\mathrm{He}_n\mathrm{V}_{m-1} + \mathrm{V} \to \mathrm{He}_n\mathrm{V}_m \quad \text{and} \quad \mathrm{He}_{n-1}\mathrm{V}_m + \mathrm{He}_{\mathrm{int}} \to \mathrm{He}_n\mathrm{V}_m$$

where He$_{\text{int}}$ indicates an interstitial tetrahedral helium atom. The values found are positive (attractive interaction) for all cases. The He binding energies are positive even in absence of vacancies ($m = 0$), the lowest energy configuration of the small He$_n$ clusters is shown in Fig. 3 (left). We also note that even small He$_n$ clusters induce large lattice distortion. Also, according to the dissociation energies shown in Fig. 3 (right), the emission of a SIA creating a vacancy is energetically more favourable than the emission of an interstitial He from a He$_n$ cluster with $n \geqslant 4$. This self-trapping of He atoms followed by the emission of a SIA may constitute the initial stage towards the formation of He bubbles observed at low temperatures in initially vacancy free lattices. For He$_n$V$_m$ clusters with a given value of m, the He binding energy decreases as the He content increases, reflecting the increase in cluster pressure caused by the accumulation of He atoms, while vacancy to cluster binding energies increase with helium content. In particular, they are always larger with than without helium. In other words, helium stabilizes vacancy-type clusters by reducing the vacancy emission rates. This is consistent with the experimental evidence that He atoms enhance the formation of nanovoids [24].

Helium atoms produced by irradiation are initially in interstitial sites of the iron matrix. According to ab initio results a fast migrating interstitial He can be deeply trapped by a vacancy and become a substitutional impurity. However, it may also find a self interstitial atom before it is trapped by a vacancy. We have therefore calculated the

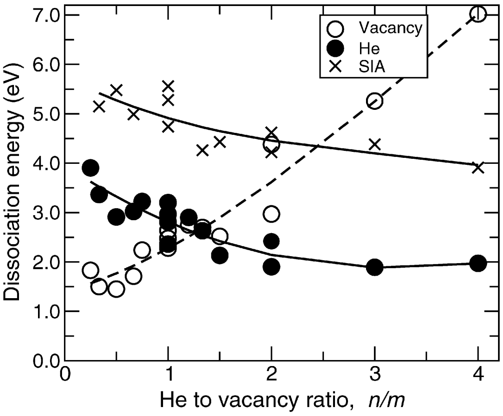

Fig. 4. Dissociation energies of a $\mathrm{He_{int}}$, a SIA or a vacancy from $\mathrm{He}_n\mathrm{V}_m$ clusters containing at least one vacancy versus He to vacancy ratio, n/m.

Fig. 4. Energies de dissociation d'un atome d'hélium interstitiel, d'un auto-interstitiel ou d'une lacune à partir d'un amas $\mathrm{He}_n\mathrm{V}_m$ contenant au moins une lacune, en fonction du raport hélium sur lacune, n/m.

interaction energies between a tetrahedral interstitial He and a $\langle 110\rangle$ dumbbell—the most stable mono-interstitial configuration—for various relative positions of the two defects. We found that several of them are attractive. The most favorable situation found is when the He atom resides in a third nearest tetrahedral site of the dumbbell. The corresponding binding energy is 0.26 eV and the dissociation energy—obtained by adding the migration energy of interstitial He—is 0.32 eV. However self-interstitial atoms can act as efficient traps for $\mathrm{He_{int}}$ only at temperatures where they are weakly mobile since their migration energy is ~0.3 eV. This will be the case only at relatively low temperatures [18].

When a self-interstitial atom approaches a substitutional He ($\mathrm{He_{sub}}$) a spontaneous recombination–replacement (kick-out) reaction is expected to occur. We confirmed that the $\mathrm{He_{sub}} + \mathrm{SIA} \rightarrow \mathrm{He_{int}}$ reaction implies a large energy gain, namely 3.6 eV. In other words the energy gained by recombining a Frenkel pair (5.9 eV) largely overcomes the energy lost by putting a helium atom from a substitutional to a tetrahedral site leaving an empty vacancy behind (2.30 eV). This reaction is responsible for the substitutional He migration by replacement mechanism activated under irradiation.

The defect ($\mathrm{He_{int}}$, SIA or V) to cluster binding energy was shown to depend strongly on the He density, i.e., the n/m ratio [19]. These clusters may change their n/m ratio by e.g. emitting He atoms, SIAs or vacancies according to their respective dissociation energies. The ab initio trends for the He (SIA, vacancy) dissociation energies from $\mathrm{He}_n\mathrm{V}_m$ were determined as a function of n/m assuming they are given by the sum of the binding energy to the cluster and of the interstitial He (SIA, vacancy) migration energy (Fig. 4). In a thermal helium desorption spectroscopy (THDS) experiment, dissociation reactions with increasing activation energies will come into play as the temperature increases. The crossover between the He and vacancy dissociation curves therefore gives information on the composition of the most stable clusters and on their stability [19]. DFT-SIESTA studies predicted the optimal n/m value to be 1.3. At variance with previous empirical potential results [19], the emission of self-interstitial atoms from clusters containing at least one vacancy always requires higher activation energies than the emission of interstitial He atoms. These ab initio results concerning the behavior of He in bcc iron have also served recently as input data for a Rate Theory model to simulate thermal desorption experiments of Vassen et al. in order to gain insights on long term evolution of He in iron [25].

4. The effects of carbon

Although carbon is present only in very small quantities in steels, it is expected to have a crucial effect on defects. Carbon interacts strongly with vacancies in iron, as attested by the radical change on the apparent vacancy formation and migration energy between ultra-pure iron, and iron containing a few appm of carbon. For this reason we have investigated the effect of carbon on the energetics of $\mathrm{He}_n\mathrm{V}_m$ helium-vacancy clusters. Some reactions selected as representative of these effects are summarized in Table 2 and the lowest energy configurations of these carbon-vacancy

Table 2
Clustering reactions involving vacancy, carbon and helium atoms. The corresponding binding energy and the dissociation energy for the respective reverse reactions are shown

Reaction	Binding energy (eV)	Dissociation energy (eV)
(1) $C + V \rightarrow VC$	0.41	1.08
(2) $VC + C \rightarrow VC_2$	0.77	1.64
(3) $V + He_{int} \rightarrow HeV$	2.30	2.36
(4) $VC + He_{int} \rightarrow HeVC$	2.09	2.15
(5) $VC_2 + He_{int} \rightarrow HeVC_2$	0.94	1.00
(4′) $HeV + C \rightarrow HeVC$	0.21	1.08
(5′) $HeVC + C \rightarrow HeVC_2$	−0.38	0.49

Fig. 5. Schematic representation of lowest energy configurations for VC and VC_2 complexes (left), and HeVC and $HeVC_2$ complexes (right). Small and large black spheres are respectively C and He atoms, grey spheres are Fe atoms, and the empty cubes represent the vacancies.

Fig. 5. Représentation schématique des configuration de plus basse énergie des complexes VC et VC2 (à gauche) et HeVC et HeVC2 (à droite). Les petites et grosses sphères noires représentent respectivement les atomes de carbone et d'hélium, les sphères grises les atomes de fer et les cubes les lacunes.

(VC, and VC_2) and helium-carbon-vacancy clusters (HeVC and $HeVC_2$) are shown in Fig. 5. The dissociation energies are calculated as the sum of the binding energy and the migration energy of the corresponding defect, where the migration energies of a vacancy, a C atom and an interstitial He are 0.67, 0.87 and 0.06 eV, respectively. The comparison between reactions (3) and (4) shows that a single carbon atom has a visible but weak effect on the binding energy between a vacancy and He_{int}, the binding energy of He_{int} with a VC complex being only 0.21 eV lower than that with a pure vacancy. On the other hand, the binding energy drops by 1.36 eV when a second carbon atom is present in the complex (reaction (5)).

The dominant mechanism for the diffusion of substitutional He, i.e. the HeV complex, under conditions with low free vacancy concentration has been suggested to be the dissociative one, as in the thermal He desorption experiment by Vassen et al. [26]. We therefore tried to estimate how the corresponding effective migration energy in pure iron, i.e. the HeV dissociation energy, could be affected by the presence of carbon. According to binding energies associated with reactions 4′ and 5′, the carbon atom will most likely weakly affect the dissociative mechanism, since the binding of carbon to HeV and HeVC is either weak or energetically unfavorable. The emission of carbon from the cluster is indeed more favorable than that of an interstitial He atom. However, recent Rate Theory simulations of Vassen's experiment clearly suggested a significant effect of impurities, most likely carbon, on the kinetics of He in bcc iron [25]. Further kinetic simulations taking explicitly into account these reactions are therefore required to give a more quantitative answer.

5. Conclusions

Ab initio studies within the framework of Density Functional Theory have been performed to study energetics and migration properties of point defects, small clusters and impurities such as He and C in α-iron. The above mentioned examples clearly showed the ability of this approach to give accurate description of defect behaviour. Thanks to the increasing computing power, combined ab initio and kinetic model simulations indeed offer new possibilities to quantitatively predict radiation damage properties in nuclear structural materials.

Acknowledgements

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