# Monte Carlo Simulation of Helium Thermal Desorption from Bcc Iron

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## ABSTRACT

Computer simulation of thermal desorption of helium pre-implanted into bcc iron has been performed using kinetic lattice Monte Carlo with *ab initio* defect interaction parameters. Quick formation of vacancy-helium clusters occurs at relatively low temperatures and limits helium release from the sample up to temperature of at least 700°C.

## 1. Introduction

Ferritic-martensitic steels accumulate in radiation environment of fusion facilities noticeable amounts of helium. At hundreds appm of implanted helium and elevated temperatures pronounced helium clustering is possible, which bears potential risk of mechanical property degradation of irradiated steels. Thus investigations of helium clustering in ferritic steels are important for prediction of He effects on the steel radiation stability.

Recent first-principles calculations of formation, binding and migration energies for vacancies, He atoms and their clusters in bcc iron have demonstrated the presence of efficient binding between He atoms and vacancies, which extends to the second nearest neighbor (NN) separations [1,2]. Such interaction creates favorable conditions for the long-range diffusion of He-V clusters via the so-called “ring” mechanism. The accelerated diffusion of small He-vacancy clusters has indeed been demonstrated in a numeric experiment [3] and it is interesting to check, whether experimental evidences in its favor can be found. One such possibility can be provided by experiments on the kinetics of He thermal desorption from He-implanted iron.

The typical scheme of experiment includes the room temperature implantation of He into iron samples up to quite high doses (so that the peak concentrations of He atoms can vary from hundreds of ppm to tens of percent), followed by annealing at the permanently increasing temperature from the ambient one to values above 1000°C. The rate of He desorption during the annealing is measured and the peaks at the desorption curve indicate the activation energies of different He release processes occurring in the annealed sample. Thus, in an experiment on thermal annealing of 8 keV He implanted iron [4] a number of peaks are observed, some of which are tentatively interpreted by the authors and some are not. Thus the lowest energy peak (at 200-250°C) is interpreted as the desorption of subsurface He, the peaks at ~500 and 750°C are attributed to the decomposition of He-rich clusters $He_nV$ (with $n > 1$) and $He_nV_m$ ($n > m > 1$), respectively, a broad peak at 800-900°C is assumed to be due to either the detrapping of He atoms from substitutional positions or to $\alpha \rightarrow \gamma$ transformation in iron, while the peak at ~1100°C has got no interpretation at all.

This tentative attribution is not very convincing. For example, the decomposition of He-rich vacancies is doubtful. The reason is not that such clusters are impossible (according to some numerical calculations, a vacancy can easily adopt two He atoms and maybe more), but the fact that each incident ion creates a lot of Frenkel pairs (~40 for the conditions of the above mentioned experiment of Sugano et al. [4], as evidenced by calculations with the code TRIM) and thus an "ovepressurized" He-V complex can easily capture sufficient amount of vacancies and decrease the chances of He emission back into the bulk. The dissociation of substitutional He into a vacancy and interstitial He requires, according to recent calculations [2], more than 2 eV and as such is also not a very efficient process even at quite high temperatures.

Some alternative mechanisms have been proposed in the literature and include in particular He transport to the sample surface along short-cuts (e.g. dislocation cores [5]) or the diffusion of small He-vacancy clusters [6]. The release peaks at high temperatures can be also associated with distinct surface damage features (such as blisters) [7]. One of these explanations is in line with our resent demonstration of accelerated diffusion of small He-V clusters. Unfortunately, it is not a trivial task to associate this mechanism with that or other desorption peak. In order to do it, one might mimic a real experimental situation with lattice kinetic Monte-Carlo (LKMC) simulations of He thermal desorption with the use of the first principles energies of He and vacancy interaction in iron, without "external" fitting parameters, and to look how closely the simulation predictions fit the experimental observations. In this paper we report some of the first results of such numerical simulations with realistic He and vacancy parameters.

## 2. Simulation details and results

Our approach to simulation of He-vacancy cluster nucleation and growth in irradiated alpha-iron, which is considered as a model material with the behavior closely resembling that of real ferritic steels, is a typical example of multiscale approach, which combines the results of quantum mechanical (*ab-initio*) calculations of He-V cluster energies with the atomistic kinetic Monte-Carlo and provides the detailed description of He diffusion and clustering that can be further dealt with at longer timescales by rougher techniques (object KMC or standard rate theory).

Simulations in this work have been performed using our Monte-Carlo code CASINO-LKMC. The code implements "continuous time" algorithm, where each Monte-Carlo step represents one successful jump and the real time per each jump is tracked. The input parameters (He and vacancy binding energies) were taken from *ab initi*o calculations [1,2], which provide the most reliable up to date estimates of defect energies. The simulation cell of 256x64x64 lattice parameters, as shown in Fig.1, is elongated in x direction (sample depth). Open boundary condition along x-direction are used, so that a vacancy or He atom reaching these cell boundaries are immediately removed. In y and z directions the boundary conditions are periodic. The initial depth distribution of He atoms, as shown in the left part of Fig.1, nearly exactly reproduced the profile predicted by TRIM calculations. Vacancy distribution follows the TRIM shape, but the peak ratio V/He was reduced to 4:1, in order to take into account the fact that typically only ~10% of atomic displacements survive the intra-cascade annealing [8]. The computational cost of simulations, which required hundreds of millions of MC steps within one run, is quite high and only several already terminated annealing runs are discussed.

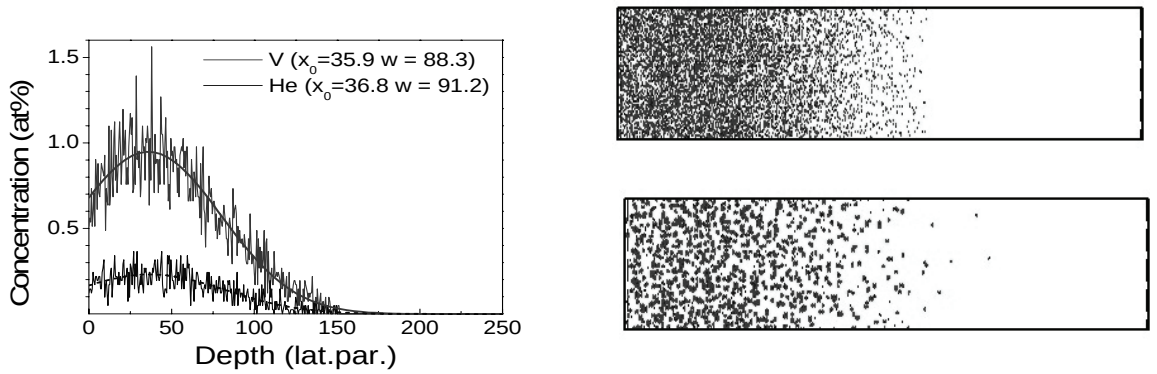
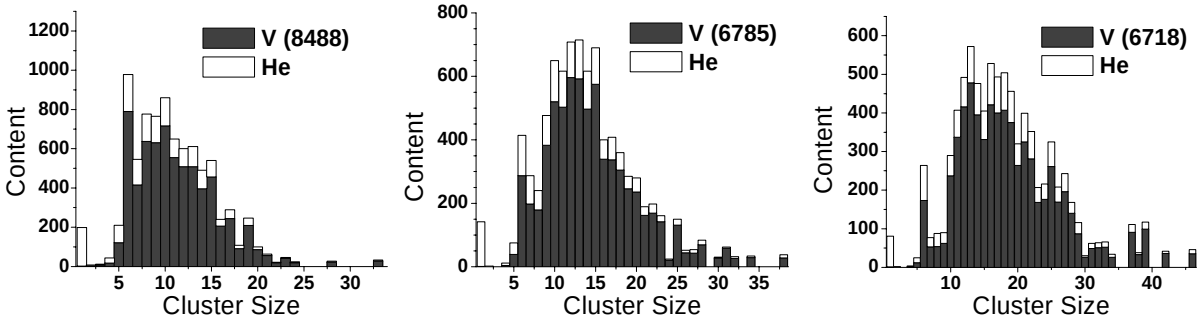



Figure 1. Left: Depth distribution of implanted He atoms (lower) and vacancies (upper curve), as predicted by TRIM for 8 keV He implantation up to $5x10^{14}$ He/cm$^2$ into iron. Right: Distribution of He atoms and vacancies in LKMC simulation cell: at the beginning of simulation (above) and after KMC annealing at 200°C for 0.2 s.

First of all, the annealing at 200°C was performed for as long as 0.2 s. By this time no vacancy monomers were left in the bulk, even though at the start of the simulation nearly all the vacancies were monomers. Very small amount of vacancies left the cell through the free surface, but the absolute majority formed clusters that contained relatively low content of He, see Fig. 2a. It is interesting to note that not only vacancy monomers, but pure vacancy clusters and He-V complexes with the sizes up to 4 disappeared (cluster size means here the number of lattice sites in the cluster, either empty of filled with He atoms). This results from the fact that such clusters are highly mobile [3] and coalesce with bigger ones.

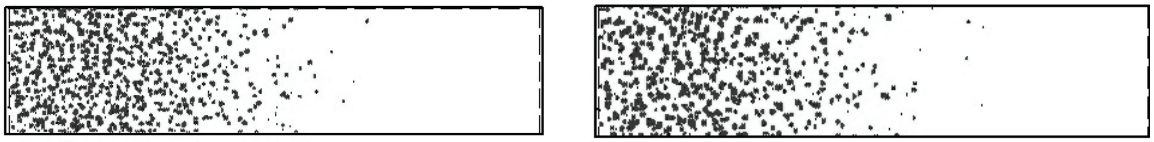



**(a)** **(b)** **(c)**

Figure 2. The content of point defects in clusters of different sizes after annealing at 200°C (a), 400°C (b) and 700°C (c). The relative parts of vacancies and He atoms within clusters of each particular size are shown in dark gray and white. The numbers in the legends indicate the total numbers of vacancies left in the simulation cell.

Only small amount of helium close to the free surface were lost. However, in real experiments the free surface is much larger than that of our simulation cell. Consequently helium leakage might be qualitatively consistent with the above mentioned interpretation of the experimentally observed weak He desorption peak at this temperature.

The final distribution of He-vacancy clusters obtained in the run at 200°C was used as a starting configuration for two runs at higher temperatures, namely at 400°C and 700°C. The final distributions of He clusters in the simulation cell are shown in Fig. 3. At both temperatures the active thermal emission of vacancies from clusters is observed, which results in the cluster ripening and the loss of vacancies at the free surface. As can be seen in the legends in Fig.2, the total loss of vacancies in both runs was comparable, which is due to the comparable numbers of MC steps at both temperatures. However, the ripening at 700°C is more pronounced, cf. Figs. 2b and 2c.



**(a)** **(b)**

Figure 3. The distribution of He-vacancy clusters in LKMC simulation cell after KMC annealing at 400°C for 1.7 ms (a) and at 700°C for 40 μs (b).

In contrast to the vacancy loss, the loss of He atoms was noticeably different. While practically no He emission was observed at 400°C, ~ 0.1% of helium atoms was lost during the run at 700°C. As can be judged from Figs.2b and c, the desorbed He involves mostly the atoms dissolved in the matrix in the form of substitutional monomers and the transport mechanism seems to be related to the formation of He-V pairs and their migration to the free sample surface.

## 3. Conclusions

He-V clustering during the linear annealing of helium ion implanted iron samples is dominated by the formation of small helium clusters at the very beginning of the temperature ramp and the ripening of the cluster ensemble at temperatures below at least 700°C. Very little part of implanted helium was lost at the surface during the KMC times reached. Yet, there is a clear trend of increasing He desorption when the temperature of 700°C is reached.

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