Work package 2 – Radiation-induced microstructure evolution

WP2 investigates how specific features of Fe-Cr-C alloys, including alloys that contain minor substitutional solutes (Ni, Si, P, …), which are model alloys for ferritic/martensitic (F/M) steels, influence and determine the microstructural changes induced in them by neutron irradiation, with a view to identifying the features that mainly cause low temperature hardening and subsequent embrittlement in F/M steels. This is done combining advanced physical models with refined multi-technique examination of irradiated materials. A comparison with ion irradiation under comparable conditions is also performed, to highlight differences or similarities between the two irradiation techniques.

The WP2 is subdivided in four tasks: The first is devoted to the study of magnetic interactions and transitions, mainly addressed by developing relevant models that account both for temperature and spin effects, with appropriate experimental verification; The second is dedicated to development of Object kinetic Monte Carlo techniques to be used to simulate the microstructure evolution under neutron irradiation, in order to provide a correct interpretation in terms of physical mechanisms; Within the third task ion and proton irradiation campaigns have been executed, in order to complement neutron irradiation that was performed previously and to provide samples to be investigated in this and other WPs; The last task focusses on post irradiation characterization of irradiated samples, and includes a variety of complementary techniques to study defect properties.

Task 2.1:Atomic-scale modelling of magnetism, finite temperature and alloys concentration effects (M1-M48)
Task leader: P.-W. Ma, UKAEA; other partners: CEA, KTH

A large quantity of results has been obtained describing thermodynamic, magnetic and alloying properties of Fe-Cr and Fe-C alloys. Density functional theory (DFT) has been the main driver for the physical descriptions, and several models that build on the DFT physics have been elaborated [1-5]. Magnetic phase transitions have been studied as functions of global and local concentration, Cr precipitation kinetics has been modelled successfully, new machine learning-based interatomic potentials have been constructed and a magnetic cluster expansion has been used to build the phase diagram of the FeCr system. Most of these results are reported in Deliverable D1.2 (D11) entitled: *Atomic-scale models for magnetism, finite temperatures and alloy concentration effects*. A vast quantity of data has been accordingly generated that is available for the project: the database has been reported in Milestone M2.2 (MS8).

Task 2.2: Modeling of microstructure evolution under neutron irradiation (M1-M43)
Task leader: N. Castin, SCK•CEN; other partners: UA

The OKMC model, developed within this task and applied to cases of interest for the project, describes the evolution of an alloy under irradiation with stochastic events implementing the migration and interaction of diffusing species, namely point defects or complexes involving point defects and solutes, e.g., vacancy-solute clusters. Key reactions in this OKMC model are the dragging of minor solute atoms by single point defects, and the binding of solute atoms with small loops, which were parameterized based on DFT calculations [6,7].

The segregation and precipitation of Cr in the presence of defects produced by the irradiation, as well as the effect of carbon impurities on the microstructure evolution are addressed by an OKMC simulation code called "MMonCa", which has been adopted and further developed in this task. In order to model the evolution of concentrated Cr solutes in Fe, a novel cellular (cell-) OKMC method was developed [8]. In this method the simulation volume is partitioned in cells. Defect migrations are biased according to the local Cr concentration in a given cell and in the neighbouring ones, following the thermodynamics of the system.

The OKMC results have been used to rationalize the experimental results of the examination of neutron irradiated Fe-Cr-C alloys by positron annihilation spectroscopy (PAS), small angle neutron scattering (SANS), and transmission electron microscopy (TEM), see [6,7], as well as the experimental resutls on ion irradiated FeCr-C alloys obtained by atom probe tomography (APT).

Task 2.3: Ion and proton irradiation of Fe-Cr-C and F/M steels (M6-M18)
Task leader: C. Heintze, HZDR; other partners: CNRS, PSI

The work within this task has been completed as reported in milestone M2.3 (MS14). Within this task, ion and proton irradiation experiments in Fe-Cr-C and F/M steels were performed. The ion and proton irradiation matrix is described and details on the individual irradiations are provided together with the milestone M2.3 (MS14). Part of the ion-irradiated samples prepared in this task complement neutron irradiated samples already available from the FP7 project MatISSE. They serve as an input to Task 2.4 "PIE of irradiated Fe-Cr samples", where they are characterized with respect to their microstructure. The other part of the samples serves as an input to WP4 and WP6.

Task 2.4: Post-irradiation examination (PIE) of irradiated Fe-Cr-samples (M1-M48)
Task leader: M.J. Konstantinovic, SCK-CEN; other partners: CNRS, HZDR, CIEMAT, IFZ

This task is devoted to the PIE of irradiated samples including neutron-irradiated Fe-Cr-C alloys, which were irradiated in the framework of the FP7/MatISSE project, together with samples of the same alloys that were ion-irradiated in **Task 2.3.**

PAS, SANS and hardness experiments were performed on all above-listed neutron irradiated materials [7]. TEM was also performed on some of the alloys[6]. The hardness measurements were found to be in excellent agreement with the tensile test results that had been published previously. They confirm the existence of irradiation hardening only in alloys that contain minor solute elements. From the microstructural point of view, both PAS and SANS experiments confirm in a consistent way the formation of vacancy clusters in all ferritic alloys [7]. These are larger in the Fe-9Cr alloy, while in alloys containing minor solutes they are smaller, but associated with solutes, suggesting that solute clusters are associated with point-defect clusters.

Most experimental microstructural results can be rationalized based on the OKMC model described above (**Task 2.2**), which is implemented in a "grey alloy" approximation only for the chromium atoms, while all other alloying elements were included explicitly. The model successfully describes the formation of NiSiP rich clusters, as observed in the experiments by PAS and SANS, predicting the correct size and density of solute-rich clusters, in good

agreement with the experimental data. Thus the model reinforces the notion that solute clusters are formed by segregation of solutes at point-defect clusters, dragged there by single point-defects. However, the predicted microstructure in terms of SIA loops is found to deviate from the experimental evidence. The two limitations of the OKMC model that could explain the deviation are: (i) lack of dislocations/grain boundaries as direct sinks present in the model, which makes it impossible to reproduce decoration and heterogeneous loop distribution; (ii) existence of a single family of the dislocation loops instead of two types of the loops with Burgers vector $a_0/2<111>$ and $a_0<100>$, correspondingly experiencing different migration energy barrier. In addition, by construction the model cannot provide information about α' precipitation. For this, the cell-OKMC (see **Task 2.2**) needs to be brought to full development and applied.

The APT has been performed on ion irradited alloy. The main results indicate that: (a) P clustering is clearly detected, (b) there is no visible sign of clustering of Ni and Si on the 3D images, but statistical treatments revealed clustering of these species together with P. Even though doubts about the capability of some techniques, e.g. CDB PAS, to unambiguously detect damage in ion irradiated materials exist, because of the limited volume of material that is affected, overall these results seem to suggest that ion irradiation is less effective than neutron irradiation in producing radiation damage, at least vacancy and solute clusters.

Magnetic effect (MAE) studies are performed on both non-irradiated and ion irradiated FeCr alloys and steels. The significant differences between Alternate Current (AC) susceptibility/MAE results in irradiated and non-irradiated samples can be interpreted in terms of partial thermal decomposition of carbides, taking place at the irradiation temperature of about 573 K. Particularly, this applies to α-Fe sample, which exhibits a clear increase of the Snoek relaxation peak (< 300 K) after irradiation. In the Fe-Cr samples, in contrast, there are either no differences between non-irradiated and irradiated samples, e.g. in Fe9Cr and Fe9CrNiSiP alloys, or there are modest reductions of the 800 K peak in irradiated Fe9Cr FM and E97 [9].

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