ADVANCES IN MULTISCALE MODELING OF MATERIAL IRRADIATION DAMAGE

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New advances and research areas in Multiscale Modeling of Material Radiation Damage are presented, considering work performed in the Universidad Politécnica Madrid, Universidad de Alicante and their collaborations. Based on the results, it can be concluded that this research is proving to be a key predictive tool. The last phase of sequential Multiscale Modeling, which corresponds to obtainment of macroscopic responses, has been started with good results in Fe and FeCu. Defect diffusion mechanisms in silicon carbide (SiC, a low activation material in nuclear fusion) can now be solved by obtaining defect energetics with a new quasi-quantum-mechanics code. The basic mechanisms of atomic displacement and threshold energies in amorphous silica (SiO_2) are being understood by using molecular dynamics, after cascade formation and types of defects were assessed in this key optical material in nuclear fusion. The study of Zr has been started as a first approach to understand effects of irradiation in ZrNb (high burnup fuels in nuclear fission), and the morphology of defects and basic diffusion data needed have been summarised.

INTRODUCCIÓN

The bases of ***Computational Multiscale Modeling*** for researching irradiation damage mechanisms in materials from their atomistic phase to the macroscopic phase were presented in Nuclear España in 2000 [1], and since then the progress and priorities of this research have been very significantly developed and systematized, with validation being linked to experiments proposed in each phase.

Its value as a predictive and alternative/complementary methodology to experiments has been confirmed, but now the existing needs for basic data to achieve this objective are defined in a much more orderly way. In some ways, this recalls the *"construction"* of nuclear databases, which have been well established for many years. The atomistic scale, connected to the microscope one, is successfully being extended to macroscopic results, and application to new non-metallic or structurally less understood materials is progressing significantly. The research and projects in the areas of Nuclear Fission, Nuclear Fusion and in the explanation of the effects that occur on Spallation Targets and Windows of accelerator-guided subcritical systems, increasingly include Multiscale Modeling of material irradiation damage with dedication of a great deal of funds and laboratories.

The number of International Projects in this area of research has very significantly increased in recent years: European Union ITEM Thematic Network for coordination of work in the area of Multiscale Modeling of irradiation damage (5th Framework Program); Integrated European Union Project SIRENA (5th Framework Program) for the study of ZrNb; International Project REVE for production of a multiscale modeling tool from the micro to the macroscopic scale that examines materials from simple metallic alloys to vessel steels; European Union Integrated Project PERFECT (6th Framework Program) for systematization of multiscale simulation models with experiments partially generated in this project and those drawn from similar projects on reactor vessel and internal steels; Spanish Project VENUS-II funded and sponsored by the *Consejo de Seguridad Nacional* (CSN) and the *Asociación de Empresas Eléctricas* (UNESA). In all these projects, both the Nuclear Fusion Institute of the UPM and the Physics Department of the University of Alicante play a leading role, while scientists from the Polytechnic University of Catalonia and from CIEMAT participate in some of them, contributing to multiscale material models and theory. The *Universidad Complutense* of Madrid and the *Universidad Carlos III* also make an experimental contribution to some of these projects.

In addition, in the development of nuclear fusion and through EFDA tasks of the EURATOM/European Union Program in the 5th and 6th Framework Programs, this research has been considered and financed in the Materials Area since 2002 as part of long-term research, with significant contributions by these Spanish groups in the areas of low- and reduced-activation ferritic-martensitic steels and the manufacturing processes for increasing their operating temperature, and the most advanced low-activation composite materials based on SiC. The National Research and Development Program of the Ministry of Science and Technology also contributes significantly to this materials fusion research.

DETERMINATION BY MULTISCALE MODELING OF THE HARDNESS IN Fe-α

This research, which has achieved successive milestones in multiscale sequencing that have already been published [2,3,4], has reached its last phase for determining the macroscopic magnitude of interest: material hardness and the stress-deformation curve [5]. It is this perspective that we intend to describe here.

Reduced-activation ferritic-martensitic steels are considered as candidate structural materials for nuclear fusion reactors because of their resistance to hardening and embrittlement at temperatures below 400°C, a working temperature that could be increased in the

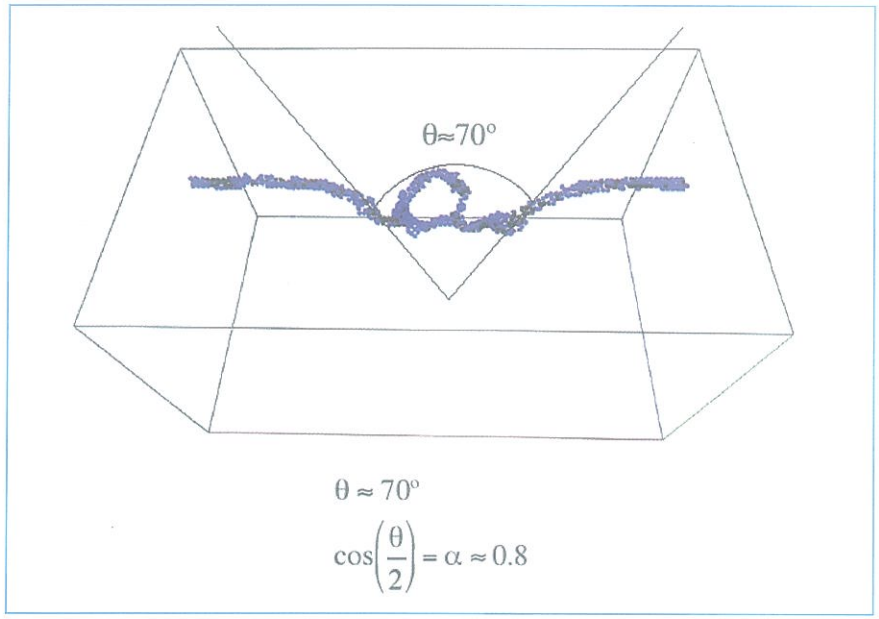

Figure 1.- Last timed image before the dislocation passes through the obstacle (defect cluster), leaving behind a loop [100] and two very curved spirals of the dislocation line. The critical angle is θ = 70°, required to estimate the resistance of obstacle α.

manufacturing process under development known as *Oxide Dispersion Strength* (ODS), in which a nano-crystalline structure is sought. Experiences with irradiated materials indicate that the existence of large dislocation loops of an interstitial nature and Burgers vector *(measure of orientation)* ½ <111> and <100> are essential for explaining the effects of hardening and embrittlement at low temperatures. Studies have been made of the effect that a high density of defect, gap and small precipitate clusters has on impeding the free movement of dislocations material. New theories have been developed, based on modeling results that describe the nature of these interstitial dislocation loops, the effect of substitutional impurities on the migration of clusters ½ <111>, and the study of the formation and growth of those of type <100> from the smallest ones ½ <111>, which appear in initial displacement cascades. These theories have helped to reconcile previous knowledge of hardening and embrittlement in Fe-α with the experiments [2, 3, 4, 5]. Although this is critical, ***what is presented in this review is the last part of the multiscale sequence*** relative to the interaction of screw-type dislocations (already existent in the original material and which are known to be responsible for control of the plastic response of bcc type materials at low temperature and under external stress) with the interstitial dislocation rings <100>. In addition to permitting a description of these mechanisms through Molecular Dynamics, **this also provides an approach to the estimation/quantification of the macroscopic response of hardness** expressed through the stress deformation curve in this material, which is consistent with *that promised* by Multiscale Modeling.

This macroscopic magnitude can be easily determined through the Orowan model that gives the change in stress $\Delta\tau_s$, $\Delta\tau_s = \alpha\ G\ b\ (Nd)^{1/2}$, as a result of the appearance of a regular set of deFects (obstacle) on the slippage plane of the dislocations. The variables that appear are: shear module (G); Burgers vector of the dislocation (b); defect density (N); defect diameter (d); and a parameter α (= cos (θ/2)) known as the *obstacle resistance* in continuous mechanics models and that is measured through the angle θ at which the dislocation is capable of overcoming the obstacle and following its path of slippage.

The shear stress originally applied was 750 MPa, and it was observed that, at this stress, the dislocation remained trapped by the obstacle [5]. Using the results obtained by Multiscale Modeling, which show that the dislocation passes the obstacle at an angle of 70° (Figure 1), meaning that the obstacle resistance is α= cos (θ/2) =0.82, and for a defect density of 1.2 x 10^{23} m^{-3}, a diameter of 2 nm and an anisotropic shear module value of 64

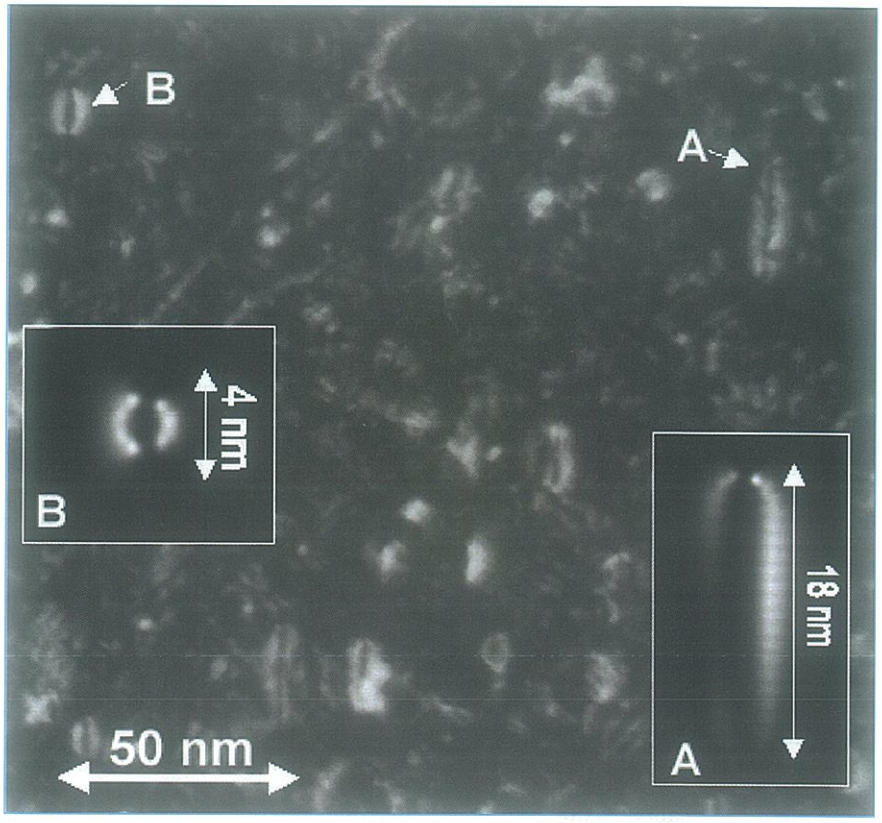

Figure 2.- Experimental TEM image of irradiated Fe-9Cr with neutrons up to 8.8 dpa at 302°C. The interior images show the result of converting via computation the multiscale modeling results (A) rectangular loop of 18 nm, (B) hexagonal loop of 4 nm, to the experimental display. Observe the pronounced agreement between experiment and simulation.

GPa, a value of $\Delta\tau_s$ = 195 MPa is determined. The values of N and d result from the type of simulation – atomistic scale of molecular dynamics – and from what is computationally admitted in it, and thus it is known they are biased for inclusion in our simulation model. The value of Dts is consequently a first approximation that can be related to the real applied stress $\Delta\sigma_u$ (=$T\Delta\tau_s$), where T is the Taylor factor with a value of 3.06, whose value would be 590 MPa. **These values, calculated for the first time by using the atomist methodology, are 50% higher than the experimental values, but for the first time they demonstrate (although with a quantitative imprecision that needs to be refined) the possibility for relating the points on the stress curve through atomistic calculations and continuous theory approximations.**

A fundamental aspect has been the ability to obtain images equivalent to visible images via Electron Microscopy (*"TEM"*), based on the results that are achieved with Multiscale Modeling on a microscopic scale. Figure 2 shows, how images obtained from our simulation results compare with visible TEM images following the technique developed by the Paul Scherrer Institute [6].

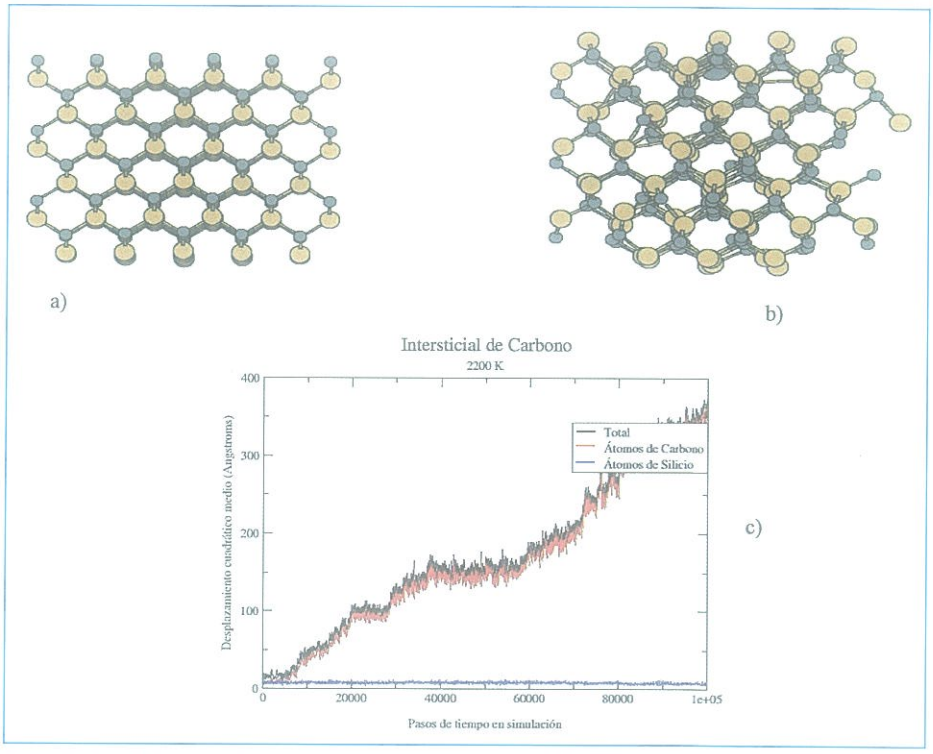

Figure 3.- Simulations using the new quasi quantum-mechanics model for SiC: a) representation of SiC crystal at temperature OK; b) structural display of SiC at 1600K; c) average quadratic displacement of a time-dependent carbon interstice in SiC, which shows the preferential displacement motion of atoms of like nature, as also occurs with Silicon through the kick out mechanism.

ENERGIES FOR DEFECT DIFFUSION IN SIC: NEW QUASI-QUANTUM-MECHANICS CODE

Silicon carbide (SiC) is a strong candidate, together with ferritic-martensitic steels and vanadium alloys, to be a structural material of the first reaction chamber wall of a nuclear fusion reactor. Structural materials must be capable of withstanding the extreme irradiation conditions in a lifetime that makes for efficient plant operation, and their rate of neutronic activation must be such that, when the plant completes its cycle (dismantling), the materials can be recycled and thus preclude storage during hundreds of years.

Research on composite materials based on SiC is developed in fusion programs of Europe/EURATOM, Japan and USA, but almost exclusively in the areas of efficient manufacturing processes and in experimental research of the consequences of irradiation in these materials (see issues of the *"Journal of Nuclear Materials"* on the "International Conference on Fusion Reactor Materials (ICFRMs)" and the "Fusion Energy Conferences" of the International Atomic Energy Agency). However, less attention is paid to the basic processes of damage to the fundamental component (SiC), and this knowledge is far from being acquired. Furthermore, while many different studies have been made on the atomistic scale and some conclusions have been reached [7, 8], progress in Multiscale Modeling has been interrupted by the shortage (non-existence) of data that allow us to advance towards the microscopic scale.

Our article in 2000 [1] explained the different multiscale methodology models and pointed out the *"perfection"* of the pure quantum-mechanics methods *("ab initio")*, together with their tremendous cost and the limitations on the size of the structures to be considered and the real time to be simulated. Other possible models were proposed which, while retaining the quantum-mechanics precision of one part of the interaction, at the same time used "some" semiempirical components, thus reducing the modeling cost. These methods, applied to the motion of atoms, are known as "tight binding molecular dynamics". A model of this kind has been successfully developed and adjusted to the SiC, so that the attractive term between system atoms receives full quantum-mechanics attention (this is the most important component of force that binds the entire structure, including electrons) and the repulsive part between them is adjusted with a semiempirical term. The primary goal has been to provide, for the first time, a tool capable of calculating formation, migration and activation energies and diffusion rates of all possible types of defects existing in SiC. This had proved to be unattainable when using classic molecular dynamics, and the only alternative was to resort to *ab initio* calculations with a tremendous computational effort. The code (TBSIC) has been successfully developed [9, 10] and checked against the basic results of the SiC structure and its dynamics. Figure 3 a) and b) shows the SiC structures in equilibrium (OK) and at a temperature of 1,600 K, for which the validity of the positions relative to the atoms in the lattice has been checked (quantitatively) through correlations of atom pairs that measure the relative distances of neighboring atoms, and their distance, to a given atom. In addition, by displaying the energy conservation over time and the binding energy of the lattice atoms, the empirical parameters of the repulsive force term have been adjusted. With this model, we have begun to determine the defect energies in SiC, as well as those of cluster formation and dissolution, with the ultimate goal of beginning a study

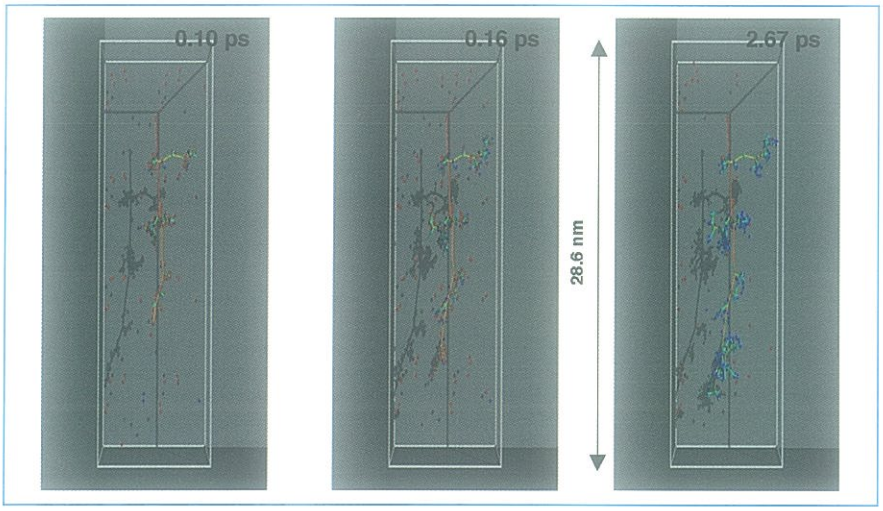

Figure 4.- Time evolution of displacement cascade formation in SiO_2 for a Primary Retrocession Atom of 5 keV. Identification of Oxygen Deficient Center defects and residual defects in the later stages.

of their diffusion and interaction on a microscopic scale and in experimentally observable periods of time, as we have already done for metals [11]. Together with these basic data, the developed model has, for the first time, made it possible to prove through simulation that the diffusion mechanism of simple interstitial defects of C and Si is the one known as "kick-out"; in this mechanism, when the interstice moves, it replaces another atom of the same nature in the lattice, causing the latter to become the new interstice. Figure 3 c) shows the evolution over time of a C interstice in SiC, and it is seen that only another atom of C is removed from its position in the lattice without any alteration of the Si atoms. The same preferential nature is observed in the case of the Si interstice and, in this case, of the Si atoms in the lattice, with the difference that the average quadratic distance covered is only 150 Å, i.e. less than C, because of the size of the C and Si atoms.

MOLECULAR DYNAMICS MODELING OF IRRADIATION DAMAGE IN AMORPHOUS SILICA

Silica (SiO_2) in its amorphous form is the primary component of most optics used in laser devices for inertial confinement fusion systems as the final focal material, and also in the heating windows of magnetic fusion equipment. Consequently, it is important to ascertain what effect radiation has on its optical properties. Significant experimental efforts have been made to prepare for operation the new demo facilities of inertial fusion ignition, i.e. the National Ignition Facility (USA), but this research has focused on the more immediate problem of ascertaining the reason for property modification when the radiation was from the laser itself being transmitted through these optics. In association with that experimental study, the first computational simulations [12] using Molecular Dynamics were made to understand the structure of the displacement cascades of this material at low energy (Figure 4). However, the current goal is to ascertain the effects of the emergent nuclear fusion neutrons and their interaction with these optic components. In previous work [13], the energy flows and spectra that could be expected in these systems were determined for a specific case of conceptual design; needless to say, these values are modified by the reactor concept. In the first phase of this research, it has been considered as essential to learn more about how the displacement of atoms in this amorphous structure occurs (i.e., know their threshold displacement energies and dependencies). By using molecular dynamics as described in [12], and with a new set of computational diagnostic models of the structure, it has been possible to begin simulating the material by causing Si and O atoms to move from low energies of 25 eV up to 200 eV and then following small energy increments and looking for the existence of stable localized defects in each case. The first necessary condition was to exactly determine that the structure being used was really amorphous silica, recognized by its typical ring morphology and different coordination number. The computational material "fabrication" procedure has been validated with the binding angle results from the experiments (Figure 5). Current results [14, 15] seem to indicate that the only threshold energy value assumed to date (117 eV) is not consistent with the reality of the fact that this threshold energy has lower values (25 – 30 eV) and is very dependent on the direction of motion of the Primary Retrocession Atom generated by the neutron.

IRRADIATION DAMAGE IN Zr / ZrNb

Understanding the irradiation damage mechanisms in ZrNb (Zr/2.5%Nb) is essential for determining whether this material can be used with full guarantees as a component in the cladding of high burnup fuel for future fission reactors. There is only a scant knowledge of the effects of irradiation in ZrNb, even on an experimental scale, at least from a theoretical-fundamental perspective. The first step being taken is to ascertain Zr behaviour (more data are available) and to develop a multiscale simulation model that extends up to the microscopeic level, making use of present knowledge about displacement cascades of this hcp material at different temperatures and generating a new database to throw light on the diffusion of defects in the material [see review in 16]. If these data are included in a new MonteCarlo diffusion model that accounts for the characteristics of Zr, they can serve to understand the microscopic evolution of Zr. The types of defects in Zr have already been recognized, the data on formation and diffusion energies available in the literature have been evaluated, and a systematic program has been developed in collaboration with the University of Liverpool to determine the parameters that are still required. Furthermore, these diffusion models include the anisotropy of defect motion in Zr, and soon it will be possible to commence the transport calculation.

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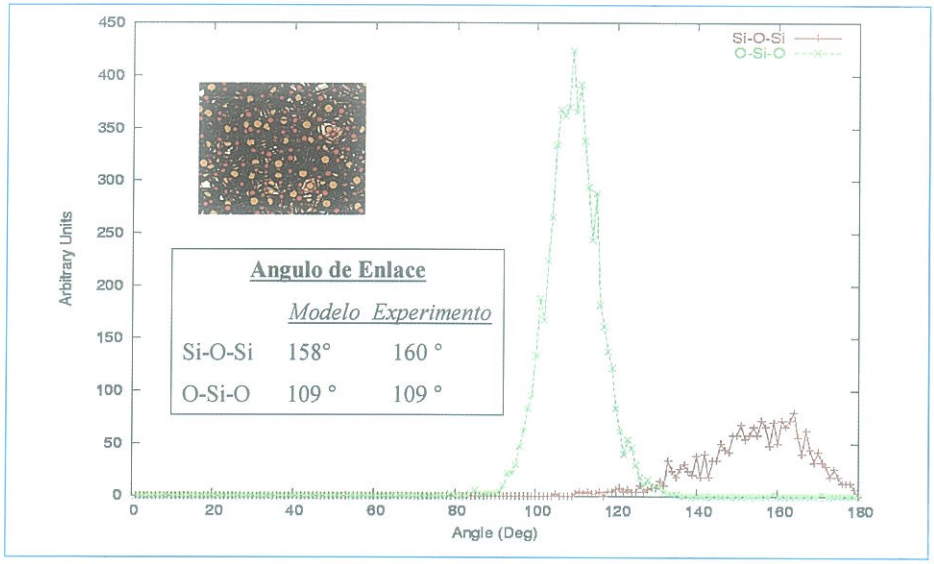

Figure 5.- Representation of the binding angles determined by Molecular Dynamics and their very good comparison with the experimental results. Indication of the correct representation of the amorphous structure of Silica via Atomistic Simulation.

Framework Program) *"Keep in Touch"*, EFDA Fusion Task / EURATOM TW3-TTMS-007 (2003), National Ministry of Science and Technology Project FTN2001-3886-C02-01 (2002-2005), and the IAEA *Coordinated Research Program on Elements of Power Plants Design for Inertial Fusion Energy* (2002-2005). The authors would like to thank Ms. Dolores Gómez Briceño and Ms. Mercedes Hernández Mayoral of *CIEMAT*, Drs. Máximo Victoria and Robin Schaublin of *l'Ecole Polytechnique Federal Lausanne/Paul Scherrer Institute*, Drs. Tomás Díaz de la Rubia and Vasily Bulatov of the *Lawrence Livermore National Laboratory*, Dr. Brian Wirth of the *University of California Berkeley*, Prof. Robert Odette of the *University of California Santa Barbara*, Prof. Luciano Colombo of *l'Universita di Cagliari*, and Prof. David Bacon of the *University of Liverpool* for all the discussions and advice on the issues addressed in this article.

REFERENCES

[1] J.M. Perlado, L. Malerba, J. Marian, "Simulación Computacional Multi-Escala del Daño inducido en los materiales por la Radiación", *Nuclear España* **194** (February 2000).

[2] J. Marian, B. D. Wirth and J. M. Perlado, *Physical Review Letters* **88** (2002) p. 255507/1-4.

[3] J. Marian, B. D. Wirth, A. Caro, B. Sadigh, G. R. Odette, J. M. Perlado and T. Díaz de la Rubia, *Physical Review* B **65** (2002) p. 094303/1-5.

[4] J. Marian, B. D. Wirth, J. M. Perlado, G. R. Odette and T. Díaz de la Rubia, *Physical Review* B **64** (2001) p. 144102/1-11.

[5] J. Marian, B. D. Wirth, R. Schäublin, G. R. Odette and J. M. Perlado, "MD modelling of defects in Fe and their interactions", accepted for publication in *Journal of Nuclear Materials*, NUMA 14404 (2003).

[6] R. Schäublin, Y. Dai, M. Victoria, *Proceedings of the 14th International Congress on Electron Microscopy*, Volume I, Eds. H. A. Calderón Benavides and M. J. Yacamán, Inst. Phy. Pub. (1998) 173-174.

[7] L. Malerba, J.M. Perlado, "Molecular dynamics simulation of irradiation-induced amorphization of cubic silicon carbide", *Journal of Nuclear Materials* **289** (2001) 57-70.

[8] L. Malerba, J.M. Perlado, "Basic mechanisms of atomic displacement production in cubic silicon carbide: a molecular dynamics study", *Physical Review* B 65 (2002) 045202/1-10.

[9] J.M. Perlado, D. Lodi, J. Marian, A. Glez. Plata and M. Salvadro, L. Colombo, M.J. Caturla and T. Diaz De La Rubia, "Time-dependent neutronics in structural materials of inertial fusion reactors and simulation of defect accumulation in pulsed Fe and SiC", *Fusion Science and Technology* **43** (2003) 384-392.

[10] J.M. Perlado, E. Domínguez, L. Malerba, J. Marian, D. Lodi, M. Salvador, E. Alonso, Ma. J. Caturla, T. Diaz De La Rubia, "Results from systematic modeling of neutron damage in inertial fusion energy reactors", *Fusion Eng. and Design* **60** (2002) 55-63, "Modelling of time-dependent damage in structural wall of inertial fusion reactors and new tight binding model for SiC", *Fus. Eng. Des.* **69** (2003) 795-801.

[11] M.Salvador, J.M. Perlado, A. Mattoni, F. Bernardini, L. Colombo, "Defects energetics of β-SiC using a new Tight Binding Molecular Dynamics Model", 11 International Conference on Fusion Reactor Materials (ICFRM), Kyoto December 2003, to be published in *Journal of Nuclear Materials* (2004).

[12] A. Kubota, M. J. Caturla, J. F. Latkowski, S. Reyes-Suarez, *Nuclear Inst. Methods* B **202** (2003) 88-92.

[13] J.M. Perlado, K. Mima, S. Nakai, E. Alonso, E. Muñoz, J. Sanz, "Activation Analysis and Materials.

Choice in the Laser Fusion Reactor *KOYO"*, *J. Nuclear Materials* **233-237** (1996) 1523-1529.

[14] J.M. Perlado, M.J. Caturla, L. Colombo, T. Díaz de la. Rubia, E. Domínguez, D. Lodi, J. Marian, F. Mota, M. Salvador, "Assessment of Structural and Silica Materials under Irradiation in Inertial Fusion Reactors: Comparison of Multiscale Modelling and Microscopy", Inertial Fusion Science and Applications (IFSA03), September 8-12, 2003 Monterey, accepted for publication in *Fusion Science and Technology*.

[15] F. Mota, M.J. Caturla, J.M. Perlado, E. Domínguez, A. Kubota, "Atomistic simulations of threshold displacement energies in SiO_2", 11 International Conference on Fusion Reactor Materials (ICFRM), Kyoto December 2003, to be published in *Journal of Nuclear Materials* (2004).

[16] C. Arévalo, J.M. Perlado, "State of art of diffusion parameters for Zr: preparing the KMC simulations",

Informe Instituto Fusión Nuclear (DENIM) / Intermediate Report EURATOM SIRENA Project March 2003.

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