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Summary

Defect production, accumulation, and materials performance in an irradiation environment

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1. Introduction

The subject of the present workshop was the defect production, accumulation, and their impact on the physical and mechanicalproperties of metals and alloys exposed to a radiation environment of energetic particles. Different aspects of these problems are treated in the individual articles of these Proceedings. Following the tradition of previous workshops in this series, a panel, composed of the present authors, was appointed to evaluate and synthesize both the overall results of the workshop and the discussion sessions.

Since the last workshop in this series, held in Montreux in 1992 (see J. Nucl. Mater. 206 (1993) 113–380), a considerable amount of progress has been made and new information has become available. Significant advances have been accomplished, for example, in the area of molecular dynamics (MD) simulations of displacement cascades in fcc and bcc metals, intermetallic compounds, and even in nonmetallic materials such as covalent semiconductors and ceramics. The simulations have provided information regarding the damage efficiency, intracascade clustering of self-interstitial atoms (SIAs) and vacancies, and the one-dimensional diffusional glide of SIA clusters. The MD simulation technique has also yielded important information regarding the stability and mobility of defect clusters formed in cascades.

Interesting progress can be also noted in the study of defect accumulation following the simulation of multiple cascades using MD. In addition a significant surge of interest in using Monte Carlo (MC) type of stochastic annealing to study cascade overlap and the long time scale evolution of the defects generated by cascades has been manifested. This is an important development since it can lead to simulations of defect accumulation which can be compared to experimental results on the microstructural evolution. Equally interesting advances have been made in the field of analytical and numerical modeling of the effects of defect accumulation on microstructural evolution under cascade damage conditions.

The challenges imposed by the need of detailed information on the physical and mechanical properties of the candidate materials for the fission as well as fusion reactors, have stimulated the community to combine several experimental methods. The progress made on the design and use of new experimental techniques provides not only a more accurate database of the macroscopical properties of the materials under an irradiation environment but also a more defined microscopical picture of the complex phenomena which occur under different irradiation conditions.

These new developments provide a very useful and exciting perspective for further in-depth study of the defect accumulation processes and their consequences in more complex systems, such as metallic alloys and ceramics. It was therefore felt that it was an appropriate time to organize a discussion meeting to explore the possibilities of further development and coordination of these new ideas and their potentials for further physical insight and understanding of radiation damage phenomena. In keeping with the tradition of this series of workshops, plenty of

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time was made available for in-depth discussions. It was hoped that the extensive and detailed nature of these discussions might help identify some of the key issues for future investigations.

2. Primary damage

The present state of computing capabilities and the use of hierarchical methods have allowed substantial progress both in the increase in PKA energy and in the extension of the temporal and spatial scales of the simulation. Thus, MD simulations using many body potential in Fe have been performed up to 40 keV, demonstrating the formation and evolution of subcascades at this energy. Cascade damage from single events to multicascade effects have been studied by combining MD with MC methods, extensively in Si but also in Cu, Fe and Au. This procedure extends the time scale of simulations from picoseconds to times in which the simulated effects can be directly compared to the experimental observation. These simulations have shown the intracascade clustering phenomena and the fact that the interstitial clusters may be glissile and extremely mobile in one dimension.

A number of presentations on the time and temperature dependent evolution of defects produced by the cascade and its comparison to the experiments emphasized the need for more understanding of defect and cluster properties in metals and alloys. It was emphasized that this will require accurate potentials that go beyond simple semi-empirical interaction forms based on effective medium concepts. It was recommended that the possibility of using first principle methods as a basis for the calculation of defect properties be explored in more detail and that these results be employed for refining the empirical potentials.

Another approach to the study of primary damage state is the so called liquid droplet model, which is based on the binary collision approximation. The model has now been refined to include heat transport in the presence of an interface (Stefan problem) and diffusion in the presence of strong thermal gradients. Yet, to make the model really predictive in terms of heat and mass transport, additional information on the liquid state of the metal is needed.

Regarding the experimental database on defect production and accumulation, the presentations on this subject made it clear that at present the community is able to extract quantitative information on some of the crucial parameters. The workshop highlighted the need for the acquisition of reliable input parameters (i.e., choice of potential, intrinsic defect properties, etc.). Effort is already in progress to determine these parameters, both in ongoing experiments and simulation calculations. The present interpretation of the experimental results indicates that only 1–10% of the defect produced by the initial cascades are available to contribute to the long range mass transport under irradiation. More detailed calculations of the sink strength in these experiments is necessary, before a definitive interpretation can provide a realistic value of the freely migrating defect fraction.

3. Defect accumulation

3.1. Modeling

Results of MC type stochastic annealing simulations of intracascade and inter-subcascade evolution of the defect population produced in cascades and subcascades were reported. Multiple-subcascades were approximated by replicating two or more MD simulated cascades in copper in several randomly chosen positions at a fixed separation distance. A group of up to five 'subcascades' of 25 keV recoil energy, was for instance studied in copper. The starting structures are taken to be the results of MD cooled cascades. The stochastic annealing simulation includes the consideration of one dimensional glide of SIA clusters present at the initial configuration. The simulation results illustrate the strong effect of the defect distribution in the primary damage state on subsequent intracascade evolution. It is found that the existence of glissile SIA clusters and the characteristics of their motion are extremely important elements in determining the nature of the global defect population. Similar results were presented in Fe where it was shown that the escape probability of defects from individual displacement cascades is of the order of 60% for both vacancies and interstitials. It was again indicated that the glide of small SIA loops plays a crucial role in determining the fate of the primary damage state over long time scale.

Further use has been made of MC techniques to study the evolution of vacancy cluster-solute complexes in RVP steels, which are shown to form in displacement cascades, and also promote Cu clustering. In Ni steels, cascades will so promote the formation of Mn–Ni-rich clusters that will harden and embrittle the steel even at low Cu contents.

The problem of global defect accumulation and microstructural evolution was treated by analytical and numerical modeling within the framework of the production bias model. The treatment is considered to be quite realistic: it takes into account the main features of the damage production in cascades, including the one-dimensional glide of the SIA clusters. The presentation reported a significant improvement in the treatment. While the earlier calculations were based on the 'mean size approximation', the new version considers the 'size distribution function' to determine various aspects of the microstructural evolution. The present approach not only provides a more accurate estimate of the microstructural parameters and their dose dependence, but also allows a proper evaluation of the impact of the glissile–sessile loop transformation on the damage accumulation behavior explicitly. The calculated void swelling values for neutron irradiated copper as a function of irradiation dose and grain size were found to be in good agreement with experimental results. The discussion on this work indicated that although the model is applicable in its present state, to pure metals, it needs to be extended to include interaction with impurities and precipitates in technological materials.

In another presentation, the phenomenon of dislocation decoration by small loops which is observed frequently under cascade damage conditions was treated in terms of segregation of three dimensionally migrating single SIAs and of one-dimensionally gliding clusters of SIAs in the vicinity of grown-in dislocations. It was shown that this phenomenon cannot be rationalized in terms of enhanced agglomeration of single SIAs, but can be understood in terms of trapping of glissile SIA loops directly produced in multi-displacement cascades. It was further shown that the irradiation-induced increase in the upper yield stress and plastic instability (i.e., yield drop) commonly observed in metals and alloys irradiated at temperatures below stage V can be understood in terms of 'cascade induced source hardening' where the grown-in dislocations are then considered to be locked by the SIA loops decorating them. It is found that the dose and temperature dependence of radiation hardening is closely related to the underlying loop accumulation near the grown-in dislocations and the stress necessary to pull a dislocation away from the 'atmosphere' of loops surrounding it.

Some effort has been devoted to establish a phenomenology for alloys under irradiation. From a theoretical point of view, an alloy under irradiation is considered as an open dissipative system which should be handled by tools other than those of classical thermodynamics. The method can be used to establish simple quantitative rules for alloy stability as a function of irradiation conditions. In another presentation, a unified thermodynamic approach to solid-state amorphization and melting was described, based on a generalized version of the Lindemann melting criterion. Within the framework of this new concept, disorder-induced amorphization is simply melting of a critically-disordered crystal at temperatures below the glass-transition point, where the supercooled liquid exists in a configurationally-frozen state, i.e., the glassy state. Evidence in support of this approach is provided by numerous experimental observations and by recent molecular-dynamics simulations of heat-induced melting and defect-induced amorphization of intermetallic compounds.

3.2. Experimental evidence

Much experimental effort has been devoted not only to observe microscopically the structure of the accumulated defects created by particle irradiation in pure and technological materials, but also to assess in extended details their contribution to the atomic transport, segregation, mechanical properties and microstructure evolution. The use of the traditional technique of observation namely transmission electron microscopy (TEM) was critically reviewed and several other techniques were introduced: scanning tunneling microscopy (STM), positron annihilation spectroscopy (PAS), etc.

To determine quantitatively the defects contributing to the atomic transport during irradiation-enhanced diffusion, a technique consisting of irradiating multi-layers, of alternating single atomic radioactive ^{60}Ni isotope and epitaxial Ni thin layers, and analyzing the concentration depth profiles with secondary ion mass spectrometry (SIMS), was presented. By using rate theory to analyze the results, the fraction of freely migrating defects (FMD) has been found to depend on temperature and the sink strength, with an FMD upper limit of 7% at 950 K. The discussion of this work demonstrated the need for combining several experimental techniques to determine quantitatively the sink density in the irradiated material and of the use of a more realistic model to be able to take into account all possible mechanisms occurring.

The PAS was presented as a sensitive technique for quantitative determination of the accumulated defects, especially the ones whose size are below the TEM resolution. It was pointed out that this technique when accompanied by other experimental investigations would be even more useful.

The STM technique is well suited for surface studies. Its application to ion irradiated nonmetals was shown to be useful in providing more insight on the interaction of bulk-surface phenomena, which hinder in many cases the success of the in situ experiments.

4. Defect production in nonmetals

Several presentations summarized fundamental aspects of defect production and accumulation in nonmetals, spanning the range from high-temperature superconductors to semiconductors and insulators. Due to the lack of reliable interatomic potentials for most semiconductors and insulators, the quantitative accuracy of the molecular dynamics simulations performed to date is uncertain. However, MD results obtained on Si and SiC provide useful insight into the experimentally-observed significant qualitative differences in their response to energetic particle irradiation. Many of the key fundamental radiation damage parameters such as threshold displacement energies, defect production efficiencies and point defect migration energies have not been accurately determined for ceramics. The potentially important role of ionizing radiation in promoting point defect diffusion in semiconductors and insulators was noted in two different presentations. In addition, experimental data on radiation-induced swelling and creep in several ceramics was reviewed in the final presentation of the workshop.

5. He and H effects: Embrittlement, swelling and creep

A further contribution to the discussion of the possible effects of He in ferritic and martensitic steels was presented, where up to 75 appm He was introduced by using different Ni isotopes in HFIR at temperatures between 300 and 600°C. At 300–400°C little effect was found of He on the mechanical properties measured up to 7 dpa, other than the hardening produced by displacement damage. At higher temperatures, actually softening is observed. In the same direction, the results of simultaneous He implantation (up to 1 at.% He) and displacement damage up to 50 dpa show that, in the temperature range up to 400°C, the hardening is mainly due to displacement damage, and that no dramatic effects of He have been found either in martensitic or 316 stainless steels.

Evidence was presented of synergistic effects between He and H in 316 l stainless steel irradiated with mixed ion beams of up to 38 keV at 573 K. The more hydrogen ions implanted, the easier and more serious is the observed blistering.

The conditions under which voids can develop in stainless steels at temperatures and displacement rates typical of PWR's, were analyzed. The results indicate that due to differences in neutron spectrum and damage rate, a 'temperature shift' would lead to more swelling per dpa than that predicted from fast reactor data. The steady state component of the creep rate is shown to be rate independent.

6. Neutron source

The requirements of materials with adequate performance in a fusion neutron environment present a pressing problem for the fusion technology community. Because of the continuing lack of a 14 MeV neutron source, the results of radiation effects studied so far are still limied to the results of computer and source simulations and irradiation with fission neutrons.

Of special interest is the present conceptual design exercise of an international fusion materials irradiation facility (IFMIF). The design is based on the D–Li reaction, which leads to a high energy recoil tail. The consensus was that, although no serious effects can be foreseen from the high energy tail of the neutron energy spectrum, there is a need for the involvement of an extended users group. As a general practical goal, the workshop addressed this problem and suggested that the necessary recommendations regarding experiment validation and analysis be performed within the fusion neutron source (IFMIF) ad hoc users' group.