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journal or publication title	Science reports of the Research Institutes, Tohoku University. Ser. A, Physics, chemistry and metallurgy
volume	35
number	2
page range	339-358
year	1991-03-05
URL	http://hdl.handle.net/10097/28353

Defect Structure Development Mechanisms during Fission and Fusion Neutron Irradiation

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(Received January 8, 1991)

Synopsis

A research history at the Oarai Branch for JMTR Utilization of the Institute of Materials Research, Tohoku University, on neutron irradiation of materials for 8 years is reviewed by quoting 35 published papers. It consists of fusion neutron irradiation with RTNS-II (Rotating Target Neutron Source, LLNL) and fission neutron irradiation with JMTR (Japan Materials Testing Reactor, JAERI).

Research program of D-T fusion neutron irradiation with RTNS-II is explained. Investigation of defect structures in a variety of materials is the basis for analysis. Major subjects are chracteristics of cascade damage, point defect processes in defect structure evolution, and recoil energy effects. By the improvement of control during JMTR irradiation, the first reliable experimental data are obtained for fission-fusion correlation. Proposals are made for the future prospect of the Oarai Branch.

I. Introduction

Irradiation study of solid materials with D-T fusion neutrons at RTNS-II, the Rotating Target Neutron Source - II at Lawrence Livermore National Laboratory, started in 1982 as a part of Japan-USA Collaboration on Fusion Research under the agreement between Monbusho (Ministry of Education, Science and Culture, Japan) and DOE (Department of Energy, USA) and continued till 1987. During five years, eighteen irradiation runs at room temperature up to 550°C with temperature interval of about 50 degrees, including several

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irradiations at low temperatures close to that of liquid helium, were performed. Maximum neutron dose attained was $6x10^{18}n/cm^2$ with three orders of different doses for each run. Each irradiation run included average of 5000 samples covering almost all conceivable materials, a large variety of metals and alloys, semiconductors and ceramics.

Although a laboratory for post-irradiation experiment was established at the site of RTNS-II, installing a high resolution electron microscope, a mechanical property test machine for miniature samples, an X-ray diffractometer etc., it was obvious that only a small portion of the required experiment could be performed there. Following a strong demand from participating experimenters, by an elaborate effort of organizing personnels, with a support from Monbusho and also with a thoughtful agreement of the Oarai Branch, a high resolution electron microscope and others were installed at the Branch particularly for the purpose of RTNS-II post-irradiation experiment. Since 1984, the experiment with samples irradiated with RTNS-II has been in progress even now after four years from the end of irradiation program.

Along the progress of post-irradiation experiment on D-T fusion neutron irradiation, the data from fission neutron irradiation to be compared were strongly desired. However, the inspection of the existing data, not only from JMTR but also from all the other reactors in the world, revealed the existence of a crucial deficiency of the control of temperature during irradiation. An in-core irradiation rig with which the sample temperature can be maintained regardless of the reactor power was designed and installed in JMTR in 1998.

As a review of the research progress, a classification of papers by quoting titles and abstracts of published papers, not newly written, is believed realistic. Details is referred to each specific paper in the references. In the last part of this review, a short remark will be made for the future progress of the research based on the experience given in the text.

II. Research Program of D-T Fusion Neutron Irradiation with RTNS-II and Preliminary Results

1. The Japanese experimental program on RTNS-II of DT-neutron irradiation of materials [1]

Research project of D-T neutron radiation damage of materials with the rotating target neutron source RTNS-II of LLNL is explained with a brief explanation of the results obtained in the first year of the project. Wide variety of materials were irradiated at three temperatures; 25°C, 200°C and 400°C up to $1\times10^{18}n/cm^2$. From the result of the observations of defect structures, discussions are made on the conditions for the defect cluster formation from cascade damage, type of defects, defects made from sub-cascade damage, correlation between defects and disordered zones, and the roles of free interstitials released from cascades. Future research plans are briefly presented.

2. Defect structure evolution from radiation damage with D-T fusion neutrons [2]

Irradiation of a variety of materials in a rotating target D-T fusion neutron source RTNS-II and observation of defect structures have been performed to obtain a unified understanding of the defect processes involved in the damage structure evolution from irradiations that generate large cascades. The maximum separation of interstitial atoms from the vacancy rich zone is measured. Vacancy type defect clusters form groups reflecting the damage with subcascades, and the three-dimensional configuration of subcascades is disclosed by high resolution stereo electron microscopy. The effective collision cross-section to produce defect clusters is estimated, and the damage efficiency is obtained.

The variation of point defect processes is discussed based on the variation of the stability of small clusters in different materials irradiated at different temperatures. The roles of free interstitials released from cascade zones, the elimination of vacancy clustered defects and the formation of their own clusters, are clearly understood from a comparison of the results of irradiation of a bulk specimen with those of thin foils. The necessity of a dynamical effect from collisions on the defect cluster formation is concluded and its mechanism is discussed. Only the descriptions are included in this paper; all the micrographs and figures will appear in another paper in the same journal.

3. Observation and analysis of defect structure evolution from radiation damage by D-T fusion neutrons [3]

A previous report on the defect structure evolution in metals, alloys and other materials by D-T fusion neutron irradiation (J. Nucl. Mater. 133&134 (1985) 85) was not accompanied with figures and illustrations, and they are all presented in this paper.

More than half of the figures consists of electron micrographs, including the following: disordered zones to show the flight distances of interstitial atoms, amorphous zones in a semiconductor, grouped defect clusters developed from sub-cascade damage, stereo-micrographs of three dimensional configuration of defect clusters in sub-cascade groups, variation of defect structures with irradiation temperature, comparison of defect structures developed in thin foil and bulk specimens to demonstrate the role of free interstitials, homogeneous and localized formation of interstitial clustered defects, detection of invisible defects by the aid of electron illumination, and dislocation structures introduced by the deformation of irradiated materials. The other figures contain numerical results of micrograph analysis, which can be used for the estimation of neutron collision cross-section and primary knock-on energy.

Point defect processes occurring during the damage structure evolution, including the dynamical effect of collisions, are discussed on the basis of experimental observations.

III. Defect Structure Evolution by D-T Fusion Neutron Irradiation in a Variety of Materials

1. Development of defect structures from displacement cascade damage in D-T neutron irradiated gold [4]

Pure gold is adopted as a suitable material for the exposure of important damage and defect processes induced by fusion neutron irradiation. Vacancy type unit defects, typically the stacking fault tetrahedra of 1-2 nm, form groups of a size up to 20 nm containing up to 20 unit defects, reflecting the damage process in the presence of sub-cascades. The three dimensional configuration of sub-cascade damage is measured and illustrated. From the density of the sub-cascade groups, the neutron collision cross-section to create the observable defects is estimated to be 1.8 barn. The observed formation of vacancy clusters at low temperatures implies the existence of the dynamical effect of collisions in the point defect reactions. The mechanism of defect structure evolution at higher temperatures, one defect cluster from one neutron collision, is discussed and the importance of point defect cluster stability and the role of free interstitials are emphasized.

2. Formation of secondary defects in copper by 14 MeV neutron irradiation and their effects on microstructure evolution [5]

Pure copper specimens were irradiated at 25, 200 and 400°C by 14 MeV neutrons using RTNS-Ii to the dose of 3.6×10^{22}n/m^2 and their damage structure was examined by means of transmission electron microscopy. At 25 and 200°C, stacking fault tetrahedra (SFT), partially dissociated Frank loops, aggregates of vacancies, and interstitial loops are nucleated by cascade collapse. They have a tendency to be formed as a group up to about 10. Because SFT are very stable under irradiation, excess interstitials corresponding to the vacancies retained in SFT are accumulated in the matrix and form their clusters. Interstitial loops nucleated near a dislocation grow preferentially by absorbing the interstitials migrating towards the dislocation. Voids were observed at 400°C. They play a very important role in void swelling at high dose.

3. Electron microscope observation of highly disordered regions in neutron irradiated germanium [6]

High purity germanium was irradiated by 14 MeV neutrons. Structural irregularities introduced at room temperature by cascade type of collision processes are of irregular shape with a faint electron microscope image contrast whose size ranges from 2 to 20 nm. High distortion of lattice images and poor stability of these damaged zones under damage sub-threshold electron illumination suggest they consist of high density distorted bonds, in an extreme a kind of amorphous zones. The collision cross section of neutrons to produce these damaged zones has been estimated to be 2.6 barns. Preliminary results on gallium antimonide are also reported.

4. Detection and analysis of microscopically invisible cascade defects in 14 MeV neutron irradiated aluminum and iron [7]

Experiments to detect damage created by cascades in aluminum and iron, in which no defect structures have been observed by electron microscopy are designed by effectively combining 14 MeV neutron irradiation with electron irradiation damage by a high voltage electron microscope.

The shrinkage and annihilation of interstitial loops formed by electron irradiation by subsequent neutron irradiation was most marked in aluminum and but occurred to some degree in iron. The result is explained by a simple model of diffusion of vacancies and interstitials based on the difference of the initial distribution profile between the two kinds of point defects in a cascade.

The extent of the annihilation of neutron radiation induced defect clusters in iron formed at room temperature is detected by the variation of the interstitial cluster formation at 323 k by high energy electron irradiation. Two annealing stages of 523-573 K and 573-623 K are observed.

5. Defect structure evolution from cascade damage in 14 MeV neutron irradiated nickel and nickel alloys [8]

D-T neutron irradiation of nickel and 4 nickel based 2 at.% alloys were performed using RTNS-II at 300 to 700 K and fluence up to $10^{23} n/m^2$. The role of free point defects was detected by the comparison of defect structures produced by irradiation of material as thin foil and those irradiated as bulk. The analysis of vacancy clusters in thin foil nickel gives the average distance between sub-cascades to be 10 nm and 14 keV as sub-cascade energy. In bulk samples interstitial loops were formed the major clustered vacancy defects in nickel at the lower temperatures were stacking fault tetrahedra, and voids at the higher temperatures. This difference reflects the variation of the stability of tetrahedra with temperature.

Under-sized solutes (Si) and extremely over-sized (Sn) enhanced the formation of loops in matrix, whereas moderately over-sized solutes (Cu, Ge) enhanced the formation of loops near dislocations. In the latter case, voids were observed since free point defects can diffuse long distances.

6. Defect structure development in 14 MeV neutron irradiated copper and copper dilute alloys [9]

Copper and twelve different copper base dilute alloys (Ni, Si, Ge and Sn) are irradiated with fusion neutrons from RTNS-II at 300-723 K, up to $6 \times 10^{22} n/m^2$. Roles of free defects are detected by the comparison of defect structures irradiated as thin foil and those as bulk, and the analysis on defect structure in pure copper gives 10 keV as sub-cascade energy.

The direct formation of vacancy clustered defects from cascade damage is enhanced by oversized solutes. Oversized solutes as well as under-sized solutes enhance the formation of the interstitial type dislocation loops by reducing the interstitial mobility and the the stabilizing the loop nucleus. Existence or non-existence of dislocation loop formation near a dislocation gives a good criteria of the bordering condition for the loop formation. Well grown voids are observed in pure copper at 563 K, but they are not formed in all the alloys of 2 at.%.

7. Positron lifetime measurement and latent vacancy clusters in 14 MeV neutron irradiated nickel [10]

The existence of electron microscopically invisible uncollapsed small vacancy clusters (latent vacancy clusters) in 14 MeV neutron irradiated Ni has been investigated by comparison between electron microscopy observation and positron lifetime measurement. In the specimen irradiated at 300 K, a long lifetime component (>300 ps) was observed. This component was not observed in specimens irradiated at 363 k but appeared again in specimens irradiated at 423 K and higher temperatures. Considering the observed defect structures (stacking fault tetrahedra and interstitial loops up to the irradiation temperature of 423 K, and voids in addition to these above 473 K), the long lifetime component at 300 K was reasonably assigned to be latent vacancy clusters and that above 423 K to be voids.

8. Confirmation of vacancy-type stacking fault tetrahedra in quenched, deformed and irradiated face-centered cubic metals [11]

An efficient and reliable method for characterizing the nature of stacking fault tetrahedra in f.c.c. metals using electron microscope diffraction image contrasts is presented. By using the 220 reflection, and thereby eliminating the contrast from overlapping stacking fault, one can differentiate between intrinsic-type stacking fault tetrahedra due to lattice vacancies and extrinsic type due to interstitial atoms. The validity of the method is examined by the observation of vacancy-type stacking fault tetrahedra in quenched metals, and by the observation of interstitial-type faulted dislocation loops. Stacking fault tetrahedra introduced by plastic deformation, electron irradiation, neutron irradiation and ion irradiation are all confirmed to be vacancy type. It was found that interstitial-type stacking fault tetrahedra do not exist.

IV. Point Defect Processes in Defect Structure Evolution by D-T Fusion Neutron Irradiation

1. Factors controlling the nature and amount of residual defects in neutron irradiated materials [12]

The requirements for progress towards a unified understanding of the defect structure evolution by high energy neutron irradiation are described.

Experimental data to be considered are categorized from the observations made on defect structures in various materials irradiated with the fusion neutron source RTNS-II. Comparison of defect structures developed in thin foils with those in bulk specimens was found to be extremely efficient in increasing our understanding of defect processes. The variation of microstructure which comes from the difference in the point defect configuration in freshly made cascade damage is discussed. The consideration of the stability of point defect clusters includes absolute instability in some materials, stacking fault tetrahedron nucleation, and cooperation among sub-cascades. Roles of interstitial atoms are evaluated, such as the annihilation within a unit cascade, free interstitial migration to eliminate vacancy clusters, and the formation of interstitial clustered defects. Dynamical effect of collisions on point defect processes is discussed. Formation of voids is explained in relation with helium production.

2. Thermal stability of cascade defects in FCC pure metals [13]

Annealing experiments on irradiated fcc pure metals have been made above room temperature to examine the thermal stability and the nature of point defect clusters formed at displacement damage cascade sites in pre-thinned specimens.

Vacancy-type loops and stacking fault tetrahedra were produced in Au irradiated with 14-MeV neutrons at temperatures higher than the stage-III temperature, and the annealing behavior of these defects was similar to that of quenched-in defects in Au.

Although the limited number of annealing experiments and quantitative TEM analyses makes it difficult to show a clear-cut interpretation, we propose the following annealing mechanism in Au, Cu, Ag and Ni irradiated with neutrons at or below the stage-III temperature. Interstitial-type defects and probably small vacancy-type defects disappear while part of submicroscopic vacancy-type defects grow and become visible at the stage-III temperature. Above this temperature, small vacancy-type defects are annealed out at higher temperatures. Some submicroscopic vacancy-type defects grew even at temperatures higher than the stage-III temperature.

3. Point defect processes in the defect structure development from cascade damage [14]

A variety of the component processes controlling the development of defect structures from large cascades have extracted from the observation of point defect clusters in 14 MeV neutron irradiated materials and alloys.

The direct formation of point defect clusters from cascades is analyzed, including the relation of damaged zone with primary knock-on energy, sub-cascade structure and formation energies, and the amount of self-annihilation of point defects within cascades. Clustered defect formation is classified from the thermal stability of small point defect clusters. Estimation of the number of free interstitials created is successfully made, and their resultant fate in the final defect structure development are categorized.

A generally applicable simplified analyses is proposed. An example is given of its application to the linear development in thin foils and the square root progress in bulk. Dynamical effect of collisions and cascade overlap effect on defect structure evolution are discussed.

4. Modification of microstructures induced by temperature variation during irradiation with 14 MeV neutrons [15]

The effect of stepwise temperature changes on the evolution of the irradiation damage microstructure in Ni which results from D-T neutron irradiation was investigated by means of transmission electron microscopy. It was shown that pre-irradiation at different temperatures strongly affects the microstructural evolution during succeeding irradiation, even if the period of the pre-irradiation was short. In the case of stepwise irradiation at 90°C/350°C, a part of the interstitial loops nucleated in the pre-irradiation at 90°C grow in the subsequent irradiation and thus suppress the formation of voids. in the case of 290°C/150°C irradiation, interstitial loops formed in the pre-irradiation disappear partly by the subsequent irradiation at 150°C.

These results show that a modification of the microstructure will occur as a result of lower temperature irradiation experiments during the start up and shut down of fission reactor operation even if the period is short.

V. Characteristics of Cascade Damage in High Energy Neutron Irradiation

1. Cascade-overlap effect on defect structure evolution revealed by repeated D-T neutron irradiation [16]

Two systematic neutron irradiation experiments have been carried out in gold and copper to investigate the effect of cascade overlap on the damage structure evolution. One was the observation of the development of defect structure as a function of irradiation dose from $10^{19}n/m^2$ to $10^{22}n/m^2$. The other was a repeated irradiation and observation, in which an identical part of the specimen was observed after several irradiations, to obtain more direct information on the formation and evolution of defect structures from displacement cascade.

Point defect clusters forming closely spaced groups, which are formed from displacement subcascades, have been confirmed. These closely spaced groups have revealed themselves by the survival of a larger number of vacancy clusters in the cascade with increasing irradiation dose by the effective annihilation of interstitials at defect clusters formed by previous irradiation.

2. Effect of cascade localization induced bias on defect structure evolution [17]

A vacancy dominant atmosphere during neutron irradiation was analyzed with special attention on the initial local distribution of point defects in a cascade, i.e. a vacancy rich region which occupies a volume smaller than that occupied by interstitials. When the diffusion of vacancies starts from this localized volume and there are sinks in the cascade, vacancies have more possibilities to be absorbed by sinks than interstitials which start from an expanded volume. Consequently the contribution of vacancies in modifying the microstructure in the cascade is larger than that of interstitials. The model was applied to experimentally observed interstitial loop shrinkage and void swelling. The importance of the effect of the difference in initial local distributions of two species of point defects in the cascade (Cascade Localization Induced Bias) was emphasized.

3. Criterion of subcascade formation in metals from atomic collision calculation [18]

Atomic collision calculation of displacement collision cascades is performed to understand the dividing process of a large cascade into subcascades, being based on the binary collision approximation under systematically varied conditions of materials and energy of incident atom. The mean distance between collisions to transfer a large kinetic energy to a target atom and the size of the vacancy rich region produced by the target atom are compared as a function of the transferred energy to a target atom. The relation between these two parameters are proposed as a criterion of subcascade formation. The criterion is applied to several metals and the tendency of the large subcascade energy for the heavier atomic species is obtained. Discussion are made on the origin of the formation of subcascade and factors which determine the subcascade parameters, such as subcascade energy and subcascade zone size.

VI. Recoil Energy Effect and Defect Processes

1. Recoil energy effects and defect processes in neutron irradiated metals [19]

Subdivision of primary recoil energy into sub-cascades in 14 MeV neutron irradiated fcc metals is analyzed in reference to the primary recoil energy spectrum, 10-20 keV per sub-cascade depending on the kind of metals. Shape and size of sub-cascade groups are also related to the recoil energy, including the anisotropy with reference to the incident neutron direction.

The extent of the overlapping of sub-cascades to produce vacancy clustered defects are classified from the spacing between sub-cascades, significant overlapping for narrow spacing less than 5 nm in Ag and Au, and no overlapping for wide separation more than 10 nm in Ni and Cu.

The amount of point defects annihilated within the cascade zone is estimated and the roles of interstitial atoms released from the zone are categorized to propose a simplified kinetics modeling of the microstructure development.

A discussion is made on the irradiation rate dependence from the defect cluster formation mechanism and from the degree of cascades-overlap.

Analysis and diagnosis of the neutron-temperature history in fission reactor irradiation leads to a proposal of an essential improvement of the reactor irradiation.

2. Recoil energy spectrum analysis and impact effect of cascade and subcascade in 14 MeV D-T fusion neutron irradiated FCC metals [20]

An analysis of cascade and subcascade formation in fcc metals by D-T fusion neutron irradiation is made by fitting the experimentally observed distribution of cascade zone size and number of subcascades to the calculated primary recoil energy spectrum. The analysis is made by categorizing subcascades into closely space (Ag, Au) and widely separated (Cu, Ni). The energy subdivided in subcascades is estimated. The estimated energy density in a subcascade is as high as several tens of eV/atom, which is high enough to raise the local temperature far beyond the melting t temperature.

A discussion is made on the reactions during cascade cooling, suggesting the inadequacy of the conventional radiation damage parameter of DPA.

The existence of impact effects from other cascades to reveal the cascade collision induced invisible vacancies as visible vacancy clusters is concluded. A kinetics type of analysis is made on the microstructure evolution by the impact effect, and the sphere of influence of the impact is estimated for each material.

3. Recoil energy spectrum effects and point defect processes characteristic of cascades [21]

A procedure is proposed for the primary recoil energy spectrum analysis of the effects of high energy particle irradiation. Comparison are made between fusion and fission neutrons. Examples of the analysis of cascades and subcascades by 14 MeV neutrons are quoted. Estimation of the deposited energy density and the concept of subcascade energy are among the major consequences. Point defect processes, including the point defect cluster formation from cascades and the reaction of freely migrating defects, are reviewed.

The importance of two kinds of point defect processes characteristic of cascade collision damage is emphasized. These are the impact effect from a cascade to point defects produced by other cascades and the effect of the difference in the localization of vacancies and interstitials in a cascade (Cascade localization Induced Bias (CLIB)).

VII. Low Temperature D-T Neutron Irradiation

Low temperature irradiation of metals and alloys was performed with a liquid helium cooled cryostat, and cryotransfer observation was performed by transferring irradiated samples to a transmission electron microscope without warming up. The cryotransfer experiment below liquid nitrogen temperature was done at LLNL, and the major part of the experiment at liquid nitrogen temperature was performed at the Oarai Branch with the irradiated samples sent back from LLNL in a liquid nitrogen container. Details of research results are reported by Y. Shimomura in this volume, and only the titles of published papers are quoted here.

1. Low temperature D-T neutron irradiation and cryotransfer observation of cascade defects of metals [22]

2. Development of defects from displacement cascade damage in low temperature D-T neutron-irradiated metals [23]

3. Cascade energy overlapping effect on defect structure development disclosed by cryo-transfer observation

of D-T neutron-irradiated metals [24]

3. 20 K cryo-transfer TEM observation of nascent displacement cascade damages in low temperature D-T neutron-irradiated metals at RTNS-II [25]

4. Cryotransfer transmission electron microscopy experiment of low-temperature 14 MeV-neutron-irradiated aluminum [26]

5. Annealing experiments of low temperature 14 MeV neutron-irradiated ordered and disordered Cu_3Au by TEM [27]

VIII. Mechanical Property Change by D-T Fusion Neutron Irradiation

Soon after the start of the irradiation program with RTNS-II, an effort started to develop the experimental technique to test mechanical properties with miniaturized samples. It had started at Hokkaido and expanded all over Japan. It included miniaturized tensile test, bulge test with 3 mm disk samples which are common to transmission electron microscope observation, and three point bending. Details of research results by the application to neutron irradiation are reported by A. Okada in this volume, and only the titles of published papers are quoted.

1. Correlation among a variety of miniaturized mechanical tests and their application to D-T neutron-irradiated metals [28]

2. Mechanical property change and its correlation with defect structure evolution in D-T neutron irradiated Au and Ni [29]

3. Mechanical property change and microstructure in fission and fusion neutron irradiated iron [30]

IX. Improvement of Control during Reactor Irradiation

1. The need for improved temperature control during reactor irradiation [31]

The irradiation conditions of materials irradiated in fission reactors are nearly always insufficiently described in the literature. A careful inspection of temperature-reactor power histories reveals a deficiency in the conventional control of irradiation temperatures. In particular, a short transient irradiation at a lower temperature commonly occurs during the startup of the reactor. A large difference in the final defect structure is expected to be caused by this transient irradiation from the mechanism of the defect structure development.

An electron irradiation with a similar transient irradiation leads to a remarkably different defect microstructure. The defect structures introduced by a fission neutron irradiation with this transient was compared with those produced by a fusion neutron irradiation with perfect temperature control without transient. The difference in structures was found to be much greater than what is normally expected between fission and fusion neutron irradiation. An essential improvement in the control of reactor irradiation is proposed.

2. Fission reactor irradiation of materials with improved control of neutron flux-temperature history [32]

Eliminating the deficiency in the conventional temperature control, irradiation of materials with the Japan Material Testing Reactor is performed with an newly-designed in-core irradiation rig with which the sample temperature can be maintained regardless of the reactor power. The defect microstructures in various materials are compared with those introduced by irradiation with conventional control.

The strong influence of the transient lower temperature irradiation during the start-up of the reactor is obvious for the samples irradiated with conventional control. The influence is fully understood from the temperature dependence of the microstructure evolution mechanism. The necessity of improved control is reconfirmed.

X. Fission-Fusion Correlation

1. Radiation rate dependence of microstructure evolution [33]

Analysis and discussion are given of the radiation rate dependence of each component reaction process in the microstructure evolution under irradiation with energetic particles.

The criteria to have radiation rate dependence are discussed from a relaxation time analysis of component phenomena (typically the time for defects to disappear to sinks) in which the overlap of component processes is the origin of the dependence. Examples for electron and neutron irradiation in at presently available facilities are presented, and the disappearance of the rate dependence by the saturation of defect structure development is pointed out.

Enhancement and suppression of the defect structure development by accelerated irradiation are categorized from the pseudo-order of reaction of each component process. A typical example of a combination of component processes is presented for the dislocation loop formation by electron irradiation with a high voltage electron microscope.

Several other sources of the rate dependence, such as cascade relaxation and the reaction of transmutation products, are pointed out.

2. Fission-fusion correlation by fission reactor irradiation with improved control [34]

The necessity of the elimination of exposure to neutrons at lower temperatures during start-up and shut-down of the reactor is confirmed by experiments which compare the result of irradiation of metals with conventional and improved temperature control in JMTR. Only several percent of exposure to neutrons at lower temperature are found to result in one hundred percent difference of radiation induced microstructures in some cases. All the differences are understood from the microstructure development mechanisms, i.e. from the temperature dependence of the stability of point defect clusters and from the position of the transient temperature along the temperature for nucleation and growth.

Fission neutron irradiation data from improved control are compared with fusion neutron irradiation data by RTNS-II. The difference of vacancy and interstitial clusters formed directly from cascades are understood when the difference in the primary recoil energy spectrum and the thermal stability of the clusters are taken into consideration. The defect structures which are developed and/or modified by the reaction of freely migration point defects, such as vacancy clusters formed from cascades in bulk samples, interstitial type dislocation structures and voids, are remarkably different between the two. Factors to be multiplied to fission neutron irradiation dose to introduce microstructures equivalent to those by fusion neutrons are found to range widely depending on the kind of microstructure, materials and irradiation temperature, from the value less than one up to 30 in the scaling of damage energy per atom.

3. Microstructure-tensile property correlation of 316 SS in low-dose neutron irradiation [35]

Annealed 316 SS was irradiated at 90 and 290°C in RTNS-II and in OWR. Radiation induced microstructures were examined by a transmission electron microscope. Very small dislocation loops of extremely high density were observed in dark field images. In the case of 90 and 290° irradiation at RTNS-II and of 90°C irradiation at OWR, the loop density increased moderately with the dpa (roughly proportional to the square root of dpa), while that of 290°C irradiation at OWR showed a stronger dpa dependence above 0.003 dpa, where the yield stress change also increased strongly. The yield strength change was roughly proportional to the square root of the defect density and was independent of the irradiation temperature and the neutron energy spectrum. The observed small dislocation loops were the origin of the yield stress change; the strength parameter was estimated to be 0.2.

XI. Proposals for Research Progress

Being based on the understanding of defect structure development by fusion and fission neutron irradiation, two categories of proposals will be made here as the summarizing remark of this review paper. proposal for the high energy neutron irradiation.

1. Advanced fission-neutron irradiation with JMTR

The present author had to spend 4 years in persuading the Oarai Branch and the JMTR the need for the irradiation with improved control, and now the third irradiation run with improved control is almost finished. The achievement of the temperature control regardless of the reactor power enabled the irradiation with intentionally cycled temperatures, and the first irradiation with cyclic temperature variation will be performed at the end of 1990 fiscal year, and the second is scheduled in 1991. However, besides of the modification of temperature controls, there are several more important tasks of JMTR and they will be briefly stated.

(a) Systematic irradiation with multi-division multi-section irradiation rig

One of the advantageous features of the irradiation with a neutron source such as RTNS-II is in the achievement of irradiations with varied irradiation dose, placing samples at different distances from the source. On the other hand, the irradiation with an atomic reactor is usually limited to a single irradiation dose by elaborate procedure and long waiting time. In order to achieve the irradiation with varied irradiation dose under controlled condition, the design of a multi-division multi-section irradiation rig is in progress. A reactor in-core irradiation rig will be divided into three to four parts, each of which can be controlled independently at different temperatures, and each division is composed of five to six sections, each of which can be removed from the reactor core during reactor operation.

In order to have a success in performing an irradiation with advanced technique, the author realize the necessity for a tight collaboration among three, the Oarai Branch of Tohoku University, JMTR of JAERI and participating experimenters. The establishment of an official active project at the Oarai Branch is strongly desired, the attitude of the JMTR for the improved irradiation technique is desired not to decelerate, and the agreement of experimenters on the necessity for the experiment should be maintained.

(b) Re-irradiation of radio-active materials

At present, any sample once exposed to neutrons is not allowed to be included for the irradiation in JMTR. The effectiveness of the neutron irradiation of samples once irradiated in different irradiation environment has been well established by the RTNS-II and JMTR irradiation by high energy electron irradiated samples. An ordinary reactor irradiation gives information only as the integrated result of the accumulated irradiation, but the re-irradiation gives information as the differentials. An official and technical procedure for re-irradiation of neutron irradiated and/or radioactive samples in JMTR is strongly desired to be established.

2. Higher fluence high-energy-neutron irradiation

A judgment for the future direction of the higher fluence high-energy-neutron irradiation experiment can be derived from the systematic study with RTNS-II and its comparison with the controlled JMTR irradiation.

(a) Up-graded RTNS-II

The major part of this review has demonstrated that the maximum dose with RTNS-II is almost entering the gate through the complex interaction among defects produced from large cascade damage, and several times more irradiation is certainly expected to disclose important aspect of heavy irradiation with real fusion neutrons.

During five years of Japan-USA collaboration on RTNS-II utilization, the feasibility to produce neutrons of five times more was realized. The utilization of up-grated RTNS-II is strongly recommended in two reasons. The one is to derive fission-fusion neutron irradiation correlation trends to a higher fluence which is inevitable for the utilization of fission reactor for fusion research. The other is to coordinate properly the larger scale of future intense neutron source.

(b) Intense high-energy-neutron source

Currently proposed intense high-energy-neutron sources use D-Li stripping reaction and produce neutrons with a broad energy spectrum. From our experience on the recoil energy spectrum analysis of fusion neutron irradiation and also from the comparison of fusion with fission, it is easily expected to encounter a comparable difficulty to convert the result from D-Li source to real fusion as the difficulty we already have in converting fission data to real fusion.

Here strongly recommended is the examination of the effect of the broad spectrum of neutron energy with an existing facility before the start of spending tremendous research money and labors. The most appropriate facility to which we can have an access is the spallation neutron source LAMPF at LANL. The LAMPF facility is announcing the acceptance of world wide users, and the scale of experiment with LAMPF is just appropriate for the present facility in the Oarai Branch. As has the Oarai Branch contributed in the fundamental study of fusion neutron irradiation with RTNS-II, so it is strongly desired to fulfill its duty in the framework of the development of reactor materials.

Acknowledgments

The part on fusion neutron irradiation was performed under the Japan-USA Fusion Cooperation Program, Collaboration on RTNS-II Utilization, sponsored by the Ministry of Education, Science and Culture, Japan. The author is grateful to Professors K. Kawamura and K. Sumita for their proper organization of the program. Post-irradiation experiment, the major part of which is the damage structure observation, has been performed with two high resolution electron microscopes (JEM 200CX) which were installed specially for the purpose of RTNS-II utilization experiment.

The part of fission reactor irradiation was performed as a research program of the Oarai Branch for JMTR Utilization of the Institute for Materials Research, Tohoku University. The author is grateful to all the members of the Oarai Branch for their assistance.

He is also indebted to the members of Materials Irradiation Division of JMTR, Oarai Research Establishment of Japan Atomic Energy Research Institute, without their cooperation this part could not be performed.

All the researches in this paper have been supported by the Grant-in-Aid of Monbusho (The Ministry of Education, Science and Culture).

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