

Conf.  
790125-29

BNL-25807

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EXPERIMENTAL EVALUATION OF THE PRIMARY DAMAGE PROCESS - NEUTRON ENERGY EFFECTS\*

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Experimental evaluation of the neutron energy dependence of the primary damage state depends upon a number of theoretical concepts. This state can only be observed after low- or perhaps ambient-temperature, low-fluence irradiations. The primary recoil energy spectrum, which determines the character of the displacement cascades, can be calculated if dosimetry has provided an accurate neutron spectrum. A review of experimental results relating neutron-energy effects shows that damage energy or damage energy cross section has often been a reliable correlation parameter for primary damage state experiments. However, the forthcoming emphasis on higher irradiation temperatures, more complex alloys and microstructural evolution has fostered a search for additional meaningful correlation parameters.

I. INTRODUCTION

The steady progress that has been made in the development of magnetic and inertial confinement devices has intensified the need for a parallel effort in materials development for fusion reactors. The basic concepts of radiation damage have been in place for more than a decade, and the experimental confirmation of many of these ideas has been achieved. Consequently, emphasis is now being placed upon understanding the evolution of microstructures from the primary damage state after long-term exposure of a solid to energetic neutrons. Experimentally, this will be a difficult task for the short and intermediate terms because we will not have intense neutron sources which produce fusion-reactor spectra during this time period. Instead, we are forced to rely upon the diverse collection of neutron sources described in the paper by Holmes and Straalsund at this conference [1]. Inasmuch as the microstructures will evolve under different spectral conditions, they will be affected by the neutron-energy dependence of the starting point called the primary damage state. Thus our immediate goal is to develop an understanding of how the primary damage state depends upon the incident neutron energy spectrum. The purpose of this paper is to review the current status of this topic. Excellent reviews of the subject have been published before [2], so that this one should not be regarded as unique or totally original. Many of the existing reviews are models of clarity and brevity and the reader should not hesitate to refer to them.

The most obvious new feature of the fusion reactor neutron spectrum is its high-energy component which spans energies from 8 to 15 MeV. Primary atom recoil spectra reflect this

component in their own high-energy components, and very energetic displacement cascades occur as a consequence. Bombardment of a solid with kilovolt heavy ions (especially self ions) can create similar high-energy cascades so that neutron-energy dependent phenomena are often investigated by this simulation method. Pertinent results of such experiments will be mentioned in this review.

The next two sections provide an introduction to the primary damage process, and the observable state(s) which derive from it. The discussion is not entirely limited to metals and alloys. Nonmetals also fulfill a role in fusion reactors, and their response to radiation can be different from that of metals. Subsequent sections deal with the theory and experiments which have been utilized to evaluate correlation parameters or to investigate the energy dependence we have been discussing. A brief discussion of some underutilized experimental techniques is included in order to stimulate the reader's own imagination. The review closes with the author's assessment of the future course of this research effort.

2. PRIMARY RECOIL SPECTRA

As stated in the Introduction, the experimental program of fusion materials development for irradiation performance will rely upon several dissimilar neutron sources. The essential difference which is relevant to intercomparison of experimental results obtained at these sources is in the primary knock-on atom energy spectrum. The size and spatial distributions of the displacement cascades that evolve from primary events depend directly on the initial kinetic energy of the recoiling atoms. This kinetic energy, corrected for the energy lost to electron excitation by all atoms comprising a cascade, is called the damage energy. Damage energy has become the most common correlation parameter now in use for comparison of results

\*Research supported by the Division of Basic Energy Sciences, Department of Energy, under Contract No. EY-76-C-02-0016.

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derived from different neutron sources. The importance of the recoil energy can be seen clearly from the results shown in the table below where recoil energy distributions and their corresponding contributions to the total damage energy are given for Nb and Cu specimens in EBRII (row 7), HFIR, a hypothetical benchmark fusion reactor first-wall spectrum (BENCH), and a "14 MeV" spectrum (actually, 13.5-15.9 MeV). A complete analysis of the results in the table can be found elsewhere [3], but it is apparent that significant differences in damage production can exist between fission and fusion spectra as well as between different types of fission reactor spectra. Examination of recoil energy groups can aid in the identification of the most significant neutron energy region of a given source spectrum. This knowledge can, in turn, facilitate the choice of a neutron or ion source for the simulation of the damage processes expected in a specific region of a fusion reactor. The kind of analysis described here has served as one basis for establishing a program of experimental evaluation of neutron energy effects.

### 3. THE PRIMARY DAMAGE PROCESS

The title of this paper is probably somewhat misleading as nobody has yet devised an experimental means of observing the primary damage process. Experimental time scales begin after the primary damage process, and at best can detect a steady-state condition in the case of low-temperature, low-fluence experiments, or some elementary microstructure which has evolved from the primary damage state owing to defect migration and interaction at elevated temperatures.

The first step in the sequence of events leading to defect formation by radiation is reasonably well understood. Most interactions of ions, electrons or neutrons with matter are described in textbooks, and need not be discussed further. The exception to this statement which is relevant here is the relative paucity of theoretical and experimental information concerning high-energy neutron scattering cross sections. More will be said about this situation in another section where the role of dosimetry in evaluation of neutron energy effects is considered in more detail.

TABLE I  
Recoil Energy, P(T), and Damage Energy Distributions,  $E_D$ , for Nb and Cu (Ref. 3)

Recoil Energy (keV)	EBRII-7		HFIR		BENCH		"14 MeV"	
	% P(T)	% $E_D$	% P(T)	% $E_D$	% P(T)	% $E_D$	% P(T)	% $E_D$
0 - 0.1	3.8	0	57.2	0	4.4	0	0.3	0
0.1 - 1.0	22.8	2.1	11.4	1.1	20.6	0.5	2.9	0
1.0 - 5.0	40.3	18.8	12.1	7.7	30.3	3.9	11.0	0.4
5.0 - 10	16.4	20.8	6.4	10.9	13.2	4.6	10.1	0.9
10 - 50	15.9	48.5	11.0	50.7	19.8	18.1	21.4	4.8
50 - 100	0.7	7.3	1.6	21.2	2.9	8.9	6.2	4.9
100 - $T_{max}$	0.1	2.5	0.3	8.4	8.8	64.0	48.1	89.0
Cu								
0 - 0.1	3.8	0	63.4	0.1	5.5	0	0.2	0
0.1 - 1.0	23.2	1.7	15.0	1.3	25.2	0.5	1.5	0
1.0 - 5.0	35.0	12.1	7.7	4.5	26.5	2.7	6.1	0.2
5.0 - 10	15.3	14.7	3.4	6.0	10.3	3.1	6.3	0.4
10 - 50	20.5	50.6	7.6	37.3	19.5	16.7	24.3	4.7
50 - 100	1.7	14.0	2.0	28.4	3.6	9.7	6.9	3.4
100 - $T_{max}$	0.5	6.9	0.9	22.4	9.4	67.3	54.7	91.3

A definition of the primary damage state is in order at this point. It is the defect configuration which exists after short-term annealing within the cascade has occurred [4]. At that time, the primary knock-on atom (PKA) has already initiated a cascade of displaced atoms, the displaced atoms have created a locally turbulent region, and the defects in that region have either annihilated, clustered or migrated from the cascade to other parts of the lattice. The goal of experiments is to determine the character of the remaining clusters and the number and type of free (i.e. unreacted) defects. This information characterizes the primary damage state.

The high-energy recoil atoms, which table 1 indicates will be for more numerous in fusion reactors than in fission reactors, produce a new feature in the primary damage state--subcascades. Calculations [3] and experiments [5-8] disclose that individual displacement cascades do not continue to evolve with ever-increasing dimensions as recoil energies increase. Instead they tend to be replaced by subcascades, and to develop "extrusions" in various crystallographic directions. Computer simulations of energetic recoils in fcc Fe reported by Beeler et al. [5] illustrate this transition from a single region of disorder for each low-energy PKA ( $\leq 10$  keV) to distinct subregions of disorder distributed along the principal trajectory line of a higher-energy ( $> 10$  keV) recoil atom. Experimental verification of this changeover in the spatial character of displacement cascades has been provided by several electron-microscope investigations, for example, those of Merkle [6], and of the Stuttgart group [7,8]. To simplify the experiments, self-ion bombardment was employed in these studies and compared with the results of exposure to neutrons. Incident ion energies were chosen to span the range over which the cascades changed appearance. Further discussion of these TEM results is deferred to section six where experimental techniques are reviewed.

From its definition, it is obvious that the primary damage state will depend upon the temperature during irradiation, the fluence and the energy spectrum of the incident particles. Experiments performed at liquid-helium temperature and low fluence are most likely to preserve the primary damage state because defect migration and cascade overlap are absent. But, on the other hand, solid and gaseous transmutation products will only be present in such low concentrations that they will not affect the development of the primary damage state. The situation is quite different at the high fluences and temperatures of technological interest. Under conditions where every atom has been displaced many times, and the original lattice has developed a complex microstructure, the processes that lead from PKAs to the primary damage state may be very different from those at low temperature. In fact it might be

asked whether a primary damage state as defined earlier still occurs in this case. The answer must be that it does, but that it is no longer frozen in for us to observe experimentally. In order to examine the influence of the microstructural changes that take place at high fluences and temperatures it will be necessary to prepare samples containing such defects and to irradiate them subsequently at low temperatures and fluences. Systematic investigations of this sort have not yet been conducted as a function of recoil energy, microstructure and temperature. However, dual ion-beam facilities are now being utilized to investigate the consequences of simultaneous helium and displacement production. More complex experiments no doubt will follow.

Unfortunately, performing irradiations and measurements at helium temperature presents many experimental problems in most cases, and the majority of experiments have been carried out at room temperature or above. This often means that the experimental apparatus is simpler, while the interpretation of results is more complicated, and care must be taken to avoid misinterpretations. Thus, low-fluences are generally employed so that cascade interactions can be avoided. This still leaves open the possibility that microstructural development within a cascade will obscure the true primary damage state in an experiment performed at elevated temperatures.

In view of the inherent difficulties in actually observing the primary damage state it seems necessary to emphasize the goals of such experiments. There are several. The foremost is to aid in the development of models which can be used to forecast radiation effects in untested radiation environments such as a proposed fusion reactor. The fruits of these investigations will be damage correlation parameters and prescriptions for describing the starting point of microstructural evolution.

While the primary damage state in metals evolves from the initial interactions encompassed by the nuclear stopping power, in nonmetals electronic losses can be important to the generation of defects. The gamma flux which always accompanies neutron irradiation further complicates analysis. It has been shown that dynamical charge-state phenomena originating under ionizing radiation conditions are sufficient to create disorder in the nonmetallic lattice. These effects and those associated with the nature of the directional bonding complicate our picture of the primary damage state in nonmetals. Nevertheless, fundamental experiments on this class of solids analogous to those on metals are required. Special materials problems in fusion reactors have been assessed recently [9], and should serve as a warning that the same kind of information about the primary damage state is needed as for metals and alloys.

#### 4. THEORETICAL BASIS FOR EVALUATING THE PRIMARY DAMAGE STATE

Experimental evaluation of the primary damage state relies heavily on theoretical concepts. These fall into two categories: those associated with the damage process and those which relate defect structures to physical property changes. Strictly speaking, the latter should not enter into the discussion. But since the choice of experimental methods is limited, those which indirectly give evidence of lattice disorder cannot be overlooked. For example, mechanical-property measurements have been made [10], but a relationship between yield stress and defect size and distribution is needed in order to link the results to the primary damage configuration.

The most frequent approach has been to use displacement rates, number of displacements, and damage energy or damage energy cross section in some combination to correlate equivalent experiments in different neutron sources. It might be worthwhile to point out here that there can be a difference between an exposure unit and a correlation parameter. The former is intended to be used as a normalization factor between experiments in arbitrary radiation sources [7]. The quantity  $\text{dpa}$  is an example of an exposure unit. It does not take into account short-term annealing, and is not necessarily expected to match experimental results. Models based upon damage energy cross sections however treat this quantity as a correlation parameter, and we shall see in a later section that in many cases the correlation has been successful. The basis for the damage energy model is not without its shortcomings however, because it is based upon the LSS theory [12] and a knowledge of nuclear cross sections which is sketchy in some cases. The LSS theory may tend to underestimate electronic losses, at least for most of the materials of interest in fission and fusion reactors. As a consequence predictions dependent upon damage energy cross sections may be untrustworthy. Therefore, one should not be surprised if experimental results cannot be correlated with such a model. Examples of this have been encountered already, and they reinforce the need for discovering additional reliable correlation parameters.

The expression below for the damage energy cross section (or specific damage energy),  $\sigma_{DE}$  illustrates the physical information needed to utilize it:

$$\sigma_{DE} = \int_0^{\infty} \sigma(E) \int_0^E \sigma_1(E) K_1(E, T) TL(T) dT dE.$$

The quantity  $TL(T)$  is the damage energy. The summation over  $l$  determines the probability that an incident neutron of energy  $E$  causes a lattice atom to recoil with energy  $T$  due to an interaction whose cross section is  $\sigma_1(E)$ . Integration over the incident spectrum  $\sigma(E)$  normalized to unit flux completes the definition. It is clear

from this definition that the damage energy cross section may not be a useful parameter for intercomparison of radiation effects in nonmetals. If prior knowledge of the importance of ionization effects is available, then it should be possible to predict the usefulness of  $\sigma_{DE}$  in such cases. Results for MgO, are an example of the successful application of  $\sigma_{DE}$ . It would not work for alkali halides, however, and other potential insulators need to be assessed in this regard when an experimental program is launched.

#### 5. ROLE OF DOSIMETRY IN EXPERIMENTAL EVALUATIONS

From the previous discussions it is evident that dosimetry plays an important role in the analysis and interpretation of experimental results. The major uncertainties occur for neutron cross sections from 10 to 40 MeV [13], the energy range of importance for quantitative interpretation of experiments conducted in (d, Be) or (d, Li) sources. With respect to the primary damage state, knowledge of gas production or transmutation product rates is not essential because the experiments are performed at low fluences. However, recoil spectra are strongly dependent upon nonelastic reactions induced by high-energy neutrons. Therefore, the development of dosimetry techniques for high-energy neutron sources has become an integral part of the overall experimental evaluation program. One of the missing links in the intercomparison of experimental results, is the availability of standardized dosimetry data. Closely related to this is the uncertainty about the required accuracy of the dosimetry information. It has been recommended that sensitivity studies be performed [14], so that their results can be used to reevaluate some of the apparent discrepancies in the literature. When we determine the sensitivity of materials damage parameters to variations in cross sections and nuclear models we can set limits on the degree of experimental uncertainty that can be tolerated in various neutron energy intervals. One of the virtues of charged-particle damage simulation is that ion beams are relatively monoenergetic and the flux can be measured quite accurately.

The solution to the dosimetry problems for high-energy neutrons will be to establish well-characterized reference spectra for 1- MeV and deuteron stripping sources. Integral testing of nuclear data at these facilities will lead to the establishment of uniform dosimetry practices by materials scientists. It is most probable that the RINS-II [15] at Lawrence Livermore Laboratory and the (d, Be) source at U.C. - Davis [16] will be selected to provide suitable reference spectra for this purpose. Multiple foil analysis, helium accumulation flux monitoring, time-of-flight and proton-recoil spectrometry can all be employed in these studies. Other techniques such as solid-state track recorders can be tested and developed concurrently. Already the differences between

time-of-flight data and multiple-foil data for a (d, Be) source [17] have revealed the significance of the low-energy component of the neutron spectrum close to the target. These results affect the damage energy calculations and hence the correlation with experiments in other neutron sources.

Two other aspects of dosimetry have a bearing on the ultimate quality of intercomparisons. First, the coexistence gamma flux affects dosimetry foils through photoneutron and photofission reactions. Measurements of gamma spectra and fluxes may be difficult in themselves but are necessary for nonmetals research. Second, special problems occur in dealing with large samples in sources which exhibit large flux gradients and rapid spatial variations in their spectrum.

In view of the gaps in our knowledge cited above, and the experimental difficulties, the agreement between measured and calculated results which will be discussed in a later section is surprisingly good. This reinforces the need for spectrum sensitivity studies so as to reduce dosimetry development to the minimum necessary.

## 6. EXPERIMENTAL TECHNIQUES FOR EVALUATION OF PRIMARY DAMAGE PROCESSES

The arsenal of available experimental techniques for investigating defects is quite large. For the purpose of studying the primary damage process or state, techniques which directly image defects are the most desirable ones. In addition, if these techniques can be employed during an irradiation at low temperature they become especially valuable. However, few of the methods in this category actually have been developed along these lines for neutron irradiations. Field ion microscopy (FIM) and transmission electron microscopy (TEM) are the two leading candidates. The latter, in particular, has long been the predominant experimental technique for qualitative and quantitative evaluation of the room-temperature cluster and cascade phenomena induced by electrons and ions as well as neutrons. The feasibility of in situ, low-temperature TEM studies of displacement cascades should be investigated. New methods of imaging and growing sophistication in the interpretation of diffraction contrast images are continual reminders of the power of the electron microscope. The recent work of Seiler et al. [7] and Jenkins and Wilkens [8] is a good example of a development in diffraction contrast theory and experiment which makes it possible to study displacement cascades in ordered alloys. Using  $\text{Cu}^+$  ions with energies from 5 to 100 keV the latter authors were able to image the disordered region centered about the displacement cascade. The sizes of the cascades were compared with the predictions of Sigmund's [13] theory and found to agree well. Alterations in cascade shape and the appearance of sub-cascades

for incident ion energies  $\geq 30$  keV were observed. Evidence for channeling effects linked to replacement sequences was also encountered. Reference to Table 1 reveals that the energy range of the  $\text{Cu}^+$  ions used nearly coincides with that of primary recoil atoms produced in proposed or available neutron sources. The information developed is thus directly applicable to the interpretation of neutron energy effects on cascade formation.

Field ion microscopy (FIM) with its inherently better resolution has been used effectively by Seidman [19] to elucidate the point-defect structure of displacement cascades. This research is complementary to the TEM work and for metals and alloys has accounted for the cascade defects which cannot be imaged in the electron microscope because they do not collapse to form dislocation loops. Comparisons of cascades from various neutron irradiations have not been reported by FIM specialists as yet. The studies are painstaking and the statistics are obviously poor, but the information that can be derived is invaluable for comparison with the predictions of computer simulation.

Electrical resistivity measurements have long been the mainstay of the radiation-damage community because they can be performed accurately and precisely. Used alone, the experimental results are subject to a variety of interpretations. But coupled with other experiments they constitute an important element of a complete investigation. The radiation-induced changes in resistivity,  $\Delta\rho$ , can be utilized along with accurate fluence measurements to determine the initial damage rates,

$$\Delta\rho/\Delta\phi = c_F c_D,$$

where  $c_F$  is the resistivity contribution per atomic fraction of Frenkel pairs, and  $c_D$  is the displacement cross section. If the number of defects produced is proportional to the damage energy, then two experimental damage rates should scale according to the spectrum averaged damage energy cross sections for the two neutron spectra involved. The resistivity changes can be used also to compare displacement efficiencies with those predicted by simple models such as a modified version of that suggested by Kinchin and Pease [20]. Conclusions drawn from these analyses can be misleading, however, and depend upon the effects of clustering on the resistivity per defect and on the accuracy of the damage energy cross sections. To some extent resistivity recovery experiments can be employed to uncover misinterpretations. The best recent example of this is in the work of Roberto et al. [21] who investigated isochronal resistivity recovery of (d, Be) neutron damage in Cu, Nb and Pt from 3 to 400 K. Although the major annealing peaks occur at the same temperatures for thermal, fission and (d, Be) neutron damage in all three elements, Roberto et al. found evidence for major configurational differences in the primary damage state between the heavier (Pt) and the

lighter (Cu, Nb) elements by comparing fission to (d, Be) damage results. The increased cascade stability for high-energy neutrons in Pt was attributed to increased clustering of interstitials. Evidently, the resistivity per defect in clusters is about equal to that for close pairs so that the damage production experiments failed to disclose an apparent mass and energy dependence of the primary damage state. Resistivity can also be utilized to detect changes in the state of order in order-disorder alloys such as alpha-brass [22]. These results can be interpreted in terms of the number of vacancies that have escaped from cascades, and subsequently enhanced equilibrium vacancy-diffusion rates at the irradiation temperature.

X-ray diffuse scattering [23] is another effective means of examining the primary damage state, and of obtaining independent information for comparison with TEM results. If highly perfect single crystals are available, diffuse intensities can be extracted from intensity measurements close to selected Bragg reflections. From these intensities the size distributions of loop-type defects can be deduced and compared with similar information derived from TEM. Post-irradiation x-ray measurements of neutron-irradiated Cu and Nb have been reported [24] for room temperature, but recent low-temperature results from Munich have not yet been published. Earlier experiments at Jülich [25] demonstrated the feasibility of such measurements at 4 K; however, being electron irradiations the experiments did not simulate neutron damage effects relevant to this discussion.

In situ internal friction experiments can furnish direct evidence of point defects escaping from cascades through the observed changes in Young's modulus. The initial rate of modulus change can be interpreted as the rate of dislocation pinning caused by the arrival of point defects at the dislocations. Goldstone and Parkin [26] carried out neutron energy dependence measurements of this kind from 2 to 24 MeV on copper samples. The results of neutron and electron irradiations were compared to determine free-interstitial production cross sections and neutron damage effectiveness. It was found that while the free-interstitial cross section increased with incident neutron energy, it remained less than one percent of the total displacement cross section. Moreover, the number of free interstitials per unit damage energy decreased with increasing neutron energy.

Other techniques which have been used to examine the energy dependence of primary damage include lattice parameter change [27], yield strength change [28], superconductor property change [29], positron annihilation [30], and the Mössbauer effect [31]. Helium accumulation flux monitors are so sensitive that helium production cross sections can be measured for relatively low fluences [32]. Although the concentration of He atoms in low fluence experiments is negligible,

attempts to observe the primary damage state at high fluences will lead to a consideration of the role of He so that knowledge of the production cross sections is important.

Finally, various other techniques can be applied to the problem. The general class of hyperfine interaction (hfi) experiments which yields information about local order has been rather neglected [33]. Mössbauer experiments have been used to some extent, but could be further exploited. Observations of the "missing recoilless fraction", for example, might yield information about void nucleation and growth as a function of neutron energy. Optical absorption and TL measurements, successfully performed during electron irradiation [34], might be similarly performed on nonmetals in appropriate ion beams. The energy spectrum of sputtered ions may be found to contain useful information on cascade "temperatures" and sizes. This will depend upon subsequent developments in the theory of sputtering. Small-angle scattering experiments (x-ray or neutron) can detect extended defects and warrant further investigation, too. Since it is generally agreed that many techniques should be brought to bear on a given problem, the methods mentioned briefly here are worthy of further consideration and will be the subject of a future workshop [35].

## 7. SUMMARY OF RESULTS OF NEUTRON ENERGY-DEPENDENCE EXPERIMENTS

Most of the experiments designed to compare the effects of fission and fusion reactor neutron irradiations have been described in previous reviews. For example, the recent paper by Wiffen and Stiegler [2] summarizes in tabular form the experiments reported prior to 1977. Their table reveals that on the basis of damage energy or damage energy cross section, the agreement between theory and experiment has been within a factor of two. Poorer agreement was found for a measurement of lattice parameter change in Mo [27] after irradiation in RTNS I. In this case the basis for comparison was the parameter, dpa; perhaps correlation with the damage energy cross section would have been more successful. The authors' conclusion concerning an increase in the ratio of interstitial clusters to free interstitials with increasing neutron energy could be tested by x-ray diffuse scattering measurements or perhaps FIM experiments.

Continued success using damage energy (or cross section) for correlation has been reported in the last two years. Initial damage production rates have been measured in Cu, Nb, Pt and in  $^{100}\text{Mo}$ ,  $^{100}\text{Mo}$  (300 ppm Ir) at 4.2 K in pure fission, fission reactor, 30- and 40-MeV (d, Be) and 14-MeV (RTNS) neutron spectra [36,37]. In all cases, scaling in terms of damage energy or damage energy cross section produced remarkably good agreement. The actual damage rates produced by 30-MeV (d, Be) neutrons were about 2/3 those produced by neutrons from the RTNS I (14.3 MeV).

The enhanced-diffusion measurements in alpha brass mentioned in the preceding section have been extended to studies of  $^{235}\text{U}$  fission-fragment damage. Guinan et al. [38] showed that the recoil spectrum from fission-fragment damage is a better approximation to a first-wall spectrum than are those produced by fission reactor (LPTR) or 14.3 MeV neutrons. The ratios of free-vacancy production in the alpha-brass samples were correctly predicted on the basis of damage energies. The use of fission fragments would appear to be an effective means of studying low-fluence effects. For high-fluences, however, the introduction of solid and gaseous fission products could create difficulties in interpretation of results.

## 8. SUMMARY AND CONCLUSIONS

A survey of experiments performed to intercompare primary damage processes in different neutron spectra indicates that damage energy cross section is a good correlation parameter. In the worst case, agreement between theory and experiment is within a factor of two. In other cases, agreement is much closer; this is especially true of the most recent experiments [21, 36, 37]. In view of the differences in the nature of the experimental measurements, in the temperatures of irradiation and in the range of neutron spectra employed the agreement seems remarkable. This remains true in spite of uncertainties in high-energy neutron cross sections which produce corresponding uncertainties in the calculated damage energy cross sections.

From the review presented in this paper the following conclusions can be drawn:

1. Damage energy is a good integral correlation parameter for experiments of the kinds thus far reported.

2. The results of irradiations in neutron spectra that span the energies used to date should be predictable on the basis of damage energy cross section for experiments of the kinds previously performed.

3. The validity of damage energy cross section as a correlation parameter for new classes of measurements and materials is unknown, and will have to be tested empirically.

4. Evaluation of the damage energy concept should continue at low temperatures for new spectra and new classes of measurements. In addition, studies at low fluences and elevated temperatures should be performed to determine how far damage energy can be pushed as a useful correlation parameter. The latter investigations should include the use of alloys as well as pure metals and of samples preconditioned by the prior addition of impurities and/or the development of microstructures.

5. The evaluation of primary damage processes should be extended to nonmetals as these special materials will play a vital role in some regions

of certain fusion reactors.

6. Damage energy cross section alone cannot provide the means for predicting microstructural development during an irradiation. New experimental techniques or more widespread use of existing techniques that are sensitive to the size and spatial distribution of defects in the primary damage state are needed. It is especially important to develop techniques that are sensitive to defect structures whose dimensions are below the practical resolution limit of the transmission electron microscope.

7. Two lines of theoretical investigation are essential to progress in predicting the effects of neutron irradiation using the primary damage state as a starting point. The first class should relate primary defect configurations to the microstructures into which they evolve. The second class should relate microstructures to physical properties.

Primary damage state experiments will continue for several years through utilization of (d, Be) sources, RTNS-II, fission reactors and ion sources. The recent utilization plan proposed for RTNS-II [39] contains a tentative list of experiments which suggests the contemporary views on low-fluence experimental methods and objectives. Nearly all of the experiments are to be conducted at 20°C or above, most will rely on the standard techniques mentioned earlier, and many are intended to continue an investigation of the primary damage process. The successes of previous investigations have undoubtedly stimulated the move toward more technologically relevant temperatures. Additional data for comparison with 14-MeV neutron results will be available from coordinated fusion materials irradiations (MFE-I, II, III) in the ORR. The continued interplay between investigations of the primary damage process and of microstructural evolution will ensure the growth of a sound fundamental data base upon which to build a successful fusion materials program.

## REFERENCES

1. J.J.Holmes and J.L.Straalsund, preceding paper.
2. See for example, F.W.Wiffen and J.O.Stiegler, Proc. 2nd Topical Meeting of the Technology of Controlled Nuclear Fusion, Richland, Washington (1976) p. 135 (CONF 760935-PI).
3. D.M.Parkin and A.N.Goland, Rad. Eff. 23 (1976) 31.
4. D.G.Doran and J.O.Schiffgens, Proc. Workshop on Correlation of Neutron and Charged Particle Damage, Oak Ridge National Laboratory (1976) p. 3 (CONF 760673).
5. J.R.Seeler, Jr., M.F.Seeler and C.V.Parks. Intern. Conf. on Radiation Effects and Tritium Technology for Fusion Reactors. ERDA/CONF 750989 (1976), p. I-362.

6. K.L.Werkle, Radiation Damage in Metals, N.L.Peterson and S.D.Harkness, editors, ASM, Metals Park, Ohio (1976) p. 58.
7. J.Seiler, M.Wilkens and K.-H. Katerbau, Proc. 3th Intern. Cong. Electron Micro., Canberra, Vol. I (1974) p. 613.
8. M.L.Jenkins, K.-H. Katerbau and M.Wilkens, Phil. Mag. 34 (1976) 1141; M.L.Jenkins and M.Wilkens, *ibid.* 1155.
9. Program Plan - Special Materials Task Group, Department of Energy, Office of Fusion Energy, Materials and Radiation Effects Branch, DOE/ET-0032/4 (1978).
10. J.B.Mitchell, Lawrence Livermore Laboratory (USA) Report, UCRL-52388 (1973).
11. See for discussion, IAEA Specialists Meeting on Radiation Damage Units in Graphite and Ferritic and Austenitic Steel, Nucl. Engrg. and Design 33 (1975) 1-90.
12. J.Lindhard, M.Scharff and M.E.Schiøtt, Kgl. Danske Videnskab. Selskab, Mat.-Sys. Medd. 33 No. 1- (1963).
13. Symposium on Neutron Cross Sections from 10 to 40 MeV, M.R.Bhat and S.Pearlstein, editors, Brookhaven National Laboratory Report BNL-NCS-50681 (July 1977).
14. Program Plan, Damage Analysis and Fundamental Studies Task Group, Department of Energy Office of Fusion Energy, Materials and Radiation Effects Branch, DOE/ET-0032/2 (1978).
15. Lawrence Livermore Laboratory Report UCRL-52000-78-3 (March 1978).
16. D.R.Nethaway, R.A.Van Konynenburg, M.W. Guinan and L.R.Greenwood, Symposium on Neutron Cross Sections from 10 to 40 MeV, M.R.Bhat and S.Pearlstein, editors, Brookhaven National Laboratory Report BNL-NCS-50681 (July 1977) p. 135.
17. D.R.Nethaway, R.A.Van Konynenburg and T.M.Adams, Lawrence Livermore Laboratory, Report UCRL-52024 (1976).
18. P.Sigmund, G.P.Scheidler and G.Roth, Proc. Conf. on Solid State Research with Accelerators, A.N.Goland, editor, Brookhaven National Laboratory Report, BNL 50083 (1968).
19. D.M.Seidman, Radiation Damage in Metals, N.L.Peterson and S.D.Harkness, editors, ASM, Metals Park, Ohio (1976) p. 23.
20. M.T.Robinson. *ibid.* p. 1.
21. J.B.Roberto, C.E.Klabunde, J.M.Williams and R.R.Coltman, Jr., J. Nucl. Mat. 73 (1978) 97.
22. A.C.Damask, R.A.Van Konynenburg, R.J.Borg and G.J.Dienes, Rad. Eff. 29 (1976) 237.
23. B.C.Larson, J. Appl. Cryst. 8 (1975) 150.
24. J.B.Roberto, J. Narayan and M.J.Saltmarsh, Proc. Intern. Conf. on Radiation Effects and Tritium Technology for Fusion Reactors, ERDA/CONF 730989 (1976) p. II-159.
25. P.Ehrhart and W.Schilling, Phys. Rev. 38 (1973) 2604; P.Ehrhart and U.Schlagheck, J. Phys. F 4 (1974) 1575; P.Ehrhard, H.-G. Haubold and W.Schilling, Adv. in Solid State Physics XIV (1974) 87.
26. J.A.Goldstone, D.M.Parkin and H.M.Simpson, Bull. Amer. Phys. Soc. 23 (1978) 239.
27. J.L.Brimhall, L.A.Charlot and H.E.Kissinger, Rad. Eff. 23 (1976) 115.
28. J.B.Mitchell, R.A.Van Konynenburg, C.J. Echer and D.M.Parkin, Proc. Intern. Conf. on Radiation Effects and Tritium Technology for Fusion Reactors ERDA/CONF 730989 (1976) p. II-172.
29. C.L.Snead, Jr., D.M.Parkin, M.W.Guinan and R.A.Van Konynenburg, Proc. 2nd Topical Meeting on the Technology of Controlled Nuclear Fusion (1977) p. 229.
30. Y.N.Platov, C.L.Snead, Jr., K.G.Lynn, A.N. Goland and M.W.Guinan (to be submitted).
31. R.J.Borg, G.J.Dienes and R.L.Lyles, Rad. Eff. 33 (1977) 105.
32. H.Farrar IV, D.W.Kneff, R.A.Britten and R.R.Heinrich, Proc. Symp. on Neutron Cross Sections from 10 to 40 MeV, M.R.Bhat and S.Pearlstein, editors, Brookhaven National Laboratory Report, BNL-NCS-50681 (July 1977) p. 175)
33. Proceedings of NATO Advanced Study Institute on Site Characterization and Aggregation of Implanted Atoms in Materials, Corsica, 10-23 September, 1978 (to be published).
34. P.W.Levy, K.Lengweiler, N.J.Carrera and P.L.Mattern, Proc. Intern. Conf. on Color Centers in Ionic Crystals, Sendai, Japan, 1974; K.J.Swyler, W.H.Hardy II and P.W.Levy, IEEE Trans. on Nucl. Sci. NS-22 (1975) 2259.
35. Workshop on Techniques for Radiation Damage Analysis, sponsored by the Office of Fusion Energy, Department of Energy, March 8-9, 1979, Oak Brook, IL (Proceedings to be published).
36. M.W.Guinan and C.E.Violet, see ref. 32.

37. M.W.Guinan and C.E.Violet (unpublished).
38. M.W.Guinan, C.E.Violet, J.H.Kinney and A.C.Damask, Lawrence Livermore Laboratory Report UCRL-81215 (August 1, 1978).
39. RTNS Utilization Plan U.S. Department of Energy Report DOE/ET-0066, September 1978.