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THE MORPHOLOGY OF COLLISION CASCADES AS A FUNCTION OF RECOIL ENERGY

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ABSTRACT

An analytical method based on defect densities has been devised to determine the threshold energies for subcascade formation in computer simulated collision cascades. Cascades generated with the binary collision code MARLOWE in Al, Cu, Ag, Au, Fe, Mo and W were analyzed to determine the threshold energy for subcascade formation, the number of subcascades per recoil per unit energy and the average spacing of subcascades. Compared on the basis of reduced damage energy, metals of the same crystal structure have subcascade thresholds at the same reduced energy. The number of subcascades per unit reduced damage energy is about the same for metals of the same crystal structure, and the average spacing of subcascades is about the same in units of lattice parameters. Comparisons between subcascade threshold energies and average recoil energies in fission and fusion neutron environments show the spectral sensitivity of the formation of subcascades.

Keywords: cascades, subcascades, recoil energy, aluminum, copper, gold, iron, molybdenum, silver, tungsten

Introduction

The interaction of high energy particles with a crystalline solid produces collision cascades, the immediate consequences of which are displaced atoms and vacant lattice sites in localized regions of the material. The configuration of a "damaged" region in the material depends on the energy of the primary recoil atom that initiates the cascade of collisions. At high recoil energies, multiple, widely-separated damage regions, commonly referred to as subcascades, can be created in a single recoil event. In this paper we examine the characteristics of subcascade production in FCC and BCC metals.

The evolution of a material's microstructure during irradiation is largely due to interaction with the point defects produced. In nascent cascades the high energy density and the close proximity of the defects lead to nearly immediate recombination of up to 70% of the defects initially produced [1]. Of the remaining point defects, only a small fraction are found to freely migrate beyond the cascade region to interact with other elements of the microstructure [2]. The spatial distribution of the initial damage, including the subscascade structure, influences the disposition of the point defects as energy dissipates from the cascade region and as subsequent thermally activated diffusion occurs.

A first step toward understanding the influence of subcascade production on microstructure evolution is determining the number, size, spacing and threshold energy for production of subcascades. The spatial distributions of point defects in high energy collision cascades have been studied in earlier investigations using binary collision computer models in Cu [3,4] and Fe [5,6]. As the graphic depictions in the earlier work demonstrate, high energy collision cascades have extremely irregular configurations. The schemes used for identifying separate damage regions are different in each of these studies, so quantitative comparisons of their results have little meaning. In the present work, quantitative information on subcascades is determined for seven FCC and BCC metals using a new approach that unambiguously defines the onset of subcascade formation.

Computations

Cascades were generated in the face-centered cubic metals Al, Cu, Ag and Au and in the body-centered cubic metals Fe, Mo and W using the binary collision computer code MARLOWE [7]. The cut-off energy was approximately the average displacement threshold energy for each material. A simulated lattice temperature of 300K was applied, and local inelastic energy losses were included. The "damage energy" of a cascade, the recoil energy minus the inelastic energy loss, is the energy available to create atomic displacements.

MARLOWE models only the collisional stage of the cascade process, which occurs in about 10^{-13} seconds. The gross spatial configuration of the cascade is determined by the location of the higher energy collisions that occur during that stage. In the present study the entities of interest are the damaged regions of the crystals, not the individual defects (although we must identify the damaged regions as collections of individual defects). Thus, MARLOWE is ideally suited to the task at hand: it models well the high energy interactions that are responsible for the overall spatial distribution of the damage with an economy that allows significant numbers of high energy cascades to be generated in a reasonable time.

In each material 30-100 recoils having randomly chosen initial directions were simulated at each of about 10 energies ranging from 1 keV to 1 MeV. The density of vacancies was determined for each cascade, and the values were averaged at each energy. The density analysis is used as a tool to determine the energies at which subcascades start to form in the various materials. The value of the density of defects depends critically on how the volume is defined. In our analysis the volume of each cascade is defined as the rectangular parallelepiped, oriented along the cubic crystal axes, that encloses the vacancy distribution. Densities by this definition are not reflective of either the very low average density of defects throughout the material or the very high average density of defects within the cores of the cascades. Little significance should be attached to the magnitudes of the values.

A different measure of defect density was used to examine the density of defects in the cascade cores as a function of recoil energy. For each cascade the average <u>local</u> density of vacancies was investigated by determining how many other vacancies on average exist within the first three neighboring shells of atoms about each vacancy in the cascade.

In each cascade, subcascades were identified using an algorithm for finding the location and size of areas of high concentrations of vacancies. The average spacing of subcascades was determined as the average of the distance of the centroid of each subcascade from that of its nearest neighbor subcascade.

Results

Figure 1 shows a plot of the cascade vacancy density within the enclosing rectangular parallelepipeds as a function of recoil energy in gold. Over the range of energies shown the densities of vacancies in the enclosing parallelepipeds vary by two orders of magnitude. In this log-log plot the density as a function of energy is well-described by two straight line segments. The energy at which the segments intersect, about 200 keV, is the energy at which subcascades begin to form regularly in gold, based on observation of graphical representations of the cascades. Similar behavior was observed in the earlier extensive graphical analysis of Cu cascades [8].

The straight line at lower energies shows that with increasing energy the single cascade region grows larger in a self-similar way (the straight line on the log-log plot is an indication of fractal behavior, albeit over a restricted range). As subcascades begin to form, the large spaces between subcascades make the cascade volume increase at a more rapid rate with energy. Thus the straight line at higher energies effectively describes the different self-similar spatial relationships of the increasing number of subcascades.

All the metals in this study exhibit a knee in the plot of vacancy density as a function of recoil energy. We identify the energy values at the knees as the threshold energies for the break-up of cascades into subcascades. This conclusion is consistent with behavior observed in a small sample of graphical representations of the cascades. The "cascade break-up energies" vary by two orders of magnitude; values for all seven metals are listed in Table 1.

Further evidence for identifying the knee in the density curve with the cascade break-up emerges when the densities are compared on the basis of reduced energy [8] rather than recoil energy. The dimensionless quantity "reduced damage energy" is the recoil damage energy divided by the value of the screened Coulomb potential at the screening radius for each metal. Thus, in each metal the energies are normalized by the strength of the atomic interaction in that metal. When compared on this basis, the knees in the plots, indicating where subcascades start to form, occur at about the same reduced recoil damage energy for each crystal structure. This is shown in Fig. 2 for the FCC metals, where the range of reduced energy of the knees, 0.050-0.070, is identified as the reduced cascade break-up energy. A similar set of curves for BCC metals reveals a reduced break-up energy range of 0.13-0.19. The reduced energy factors and the reduced damage energies at break-up are listed in Table 1.

The steep decrease in vacancy density with increasing energy exhibited in Figs. 1 and 2 does not mean that the local density of vacancies in the core of the cascade is changing with energy, even at low energies. In earlier work [9] the average local density of vacancies in cascades in Cu was investigated. It was found for copper that this value is constant at energies above a few keV, well below the threshold for subcascade production. Computer simulations of short-term annealing of cascades in copper [10] also showed that the post-annealing numbers of residual defects and freely migrating defects increases linearly with damage energy in high energy cascades. This implies that the average environment in the vicinity of a defect, which will most strongly influence defect interactions, does not change much with energy. Constant local vacancy densities are also observed in all the FCC and BCC metals investigated here. Table 1 lists the values of local vacancy density in high energy cascades (i.e., where local vacancy densities are constant) for all the metals. Even though the reduced break-up energies are about the same within the same crystal structure, the average local densities increase approximately linearly with atomic number.

Subcascade identification was done only for Cu, Au, Fe and W. The number of subcascades produced increases linearly with recoil damage energy. Table 2 contains the average number of subcascades per recoil per keV of damage energy, the average damage energy per subcascade, and the average separation of subcascades. In each material at each energy the distribution of subcascade spacing is very broad, having a standard deviation of about +/- 100% of the mean value. Thus any differences between the metals should be considered small, and we conclude that the average spacing of subcascades is about the same in all the metals.

Discussion

Within the same crystal structure, the subcascade formation characteristics appear to be about the same in all metals when compared on the basis of the reduced energy (Fig 2). This is true only for the gross spatial distribution aspects of the cascades, which are governed by the high energy collisions. The average local densities of defects in the cascade cores are different for each metal (Table 1), showing a dependence on atomic number (or atomic mass). The differences in local defect densities, and the implied differences in local energy densities, will surely lead to differences in disposition of the defects subsequent to the collisional phase.

There is much experimental evidence purporting to support the concept of subcascades, especially the obvious grouping of clusters or loops seen in micrographs after low fluence irradiations [e.g. 11 and 12]. However, micrographs show the structures that exist long after the cascade has occurred. Another important consideration when attempting to discern subcascades in observed damage in different metals is that for each metal the configuration of cascades in a given irradiation environment depends on the energy spectrum of recoil atoms produced by the irradiating particles. The recoil spectrum is a strong function of atomic number. Figures 3A and 3B show the spectrally averaged recoil energies of FCC and BCC metals, respectively, produced by 14 MeV neutrons and by the neutrons of a typical fast reactor. The average recoil energies, displayed here in terms of reduced damage energy, vary over several orders of magnitude. The maximum recoil energies may be up to a factor of ten higher than the average energies. The cascade break-up energies, constant for each crystal structure, are shown for comparison. Thus, irradiation of Cu with 14 MeV neutrons produces mostly cascades having subcascades, while irradiation of Au with 14 MeV neutrons produces mostly single cascades. In a fast reactor (or reactors with even softer spectra) few cascades in either metal will have subcascades.

According to Fig 3A, one should observe almost exclusively single cascades in Au irradiated with 14 MeV neutrons. Micrographs of 14 MeV neutron-irradiated Au show damage in the form of groups of closely spaced dislocation loops, which have been interpreted [11,12] as forming from subcascades. These observations are not necessarily inconsistent with our analysis, which deals with clearly defined, widely spaced subcascades. However, within single simulated cascades in gold we have observed separate regions of higher vacancy density, indicating perhaps subcascades that are simply next to each other. The concept of cascade "lobes," i.e., contiguous but distinct damage regions, was developed in the earlier graphical analysis of computer simulated cascades in Cu [3].

The experimentally measured average spacings of damage regions in 14 MeV neutron-irradiated Cu and Au are reported to be 12 nm and 4 nm respectively [12]. The value for Cu is consistent with our modeling results for subcascades. The close spacing of the observed damage regions in Au would suggest that they may each be the residue of a collapsed lobe rather than the residue of subcascades by the conventional definition. An alternative viewpoint is that the residue of a single subcascade may well consist of more than one visible object and may include both vacancy and interstitial components.

Conclusions

An analytical method based on defect densities has been devised to determine the threshold energies for subcascade formation in computer simulated cascades. Compared on the basis of reduced damage energy, metals of the same crystal structure have subcascade thresholds at the same reduced damage energy.

Average recoil energies in the various metals may be above or below the thresholds for subcascade production, depending on the irradiation environment. In Cu and Fe (and, by inference, in steels and copper alloys), fusion neutrons will generally produce many subcascades while fission reactor neutrons will produce few.

The average local defect density within the core of MARLOWE cascades is constant above a minimum energy that is lower than the subcascade threshold energy. The average local vacancy density increases approximately linearly with the atomic number of the metal, which will affect the subsequent development of the damage regions in the various metals.

The number of subcascades per unit reduced damage energy is about the same for all metals of the same crystal structure. The average spacing of subcascades is about 40 lattice parameters in Cu, Au, Fe and W.

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TABLE 1

Cascade Break-up Energies and Local Vacancy Densities

Element	Reduced Energy Factor[a] (eV^{-1})	Cascade Break-up Recoil Energy (keV)	Reduced Break-up Damage Energy	Local Vacancy Density[b] (%)
Al	2.34×10^{-5}	2.5	0.052	4.2
Cu	3.05×10^{-6}	20	0.049	6.7
Ag	1.28×10^{-6}	50	0.055	7.4
Au	4.18×10^{-7}	200	0.071	13.3
Fe	4.00×10^{-6}	50	0.15	7.5
Mo	1.80×10^{-6}	130	0.19	9.2
W	5.10×10^{-7}	300	0.13	10.2

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[a] for screening radii used in the MARLOWE calculations

[b] per cent of sites within three nearest neighbors that contain vacancies

TABLE 2

Average Number and Separation of Subcascades

Element	Subcascades/PKA/ Damage Energy (keV)	Damage Energy/ Subcascade (keV)	Average Separation (lattice param.)	(nm)
Cu	0.040	25	36	13
Au	0.004	250	36	14
Fe	0.030	33	54	15
W	0.004	250	36	11

Figure Captions

1. Average Density of Vacancies in Collision Cascades in Gold as a Function of Recoil Energy. The volume of each cascade is taken as the rectangular parallelepiped oriented along the crystal axes enclosing the vacancy distribution of the cascade.

2. Average Density of Vacancies in Collision Cascades in FCC Metals as a Function of Reduced Recoil Damage Energy. The cascade break-up energy is defined as the region of reduced recoil damage energy where the slopes of the curves change.

3. Average Reduced Recoil Damage Energies in a) FCC and b) BCC Metals Irradiated with 14 MeV Neutrons and Fast Reactor Neutrons. The cascade break-up energies are shown for comparison.

Average Density of Vacancies in Collision Cascades in Gold

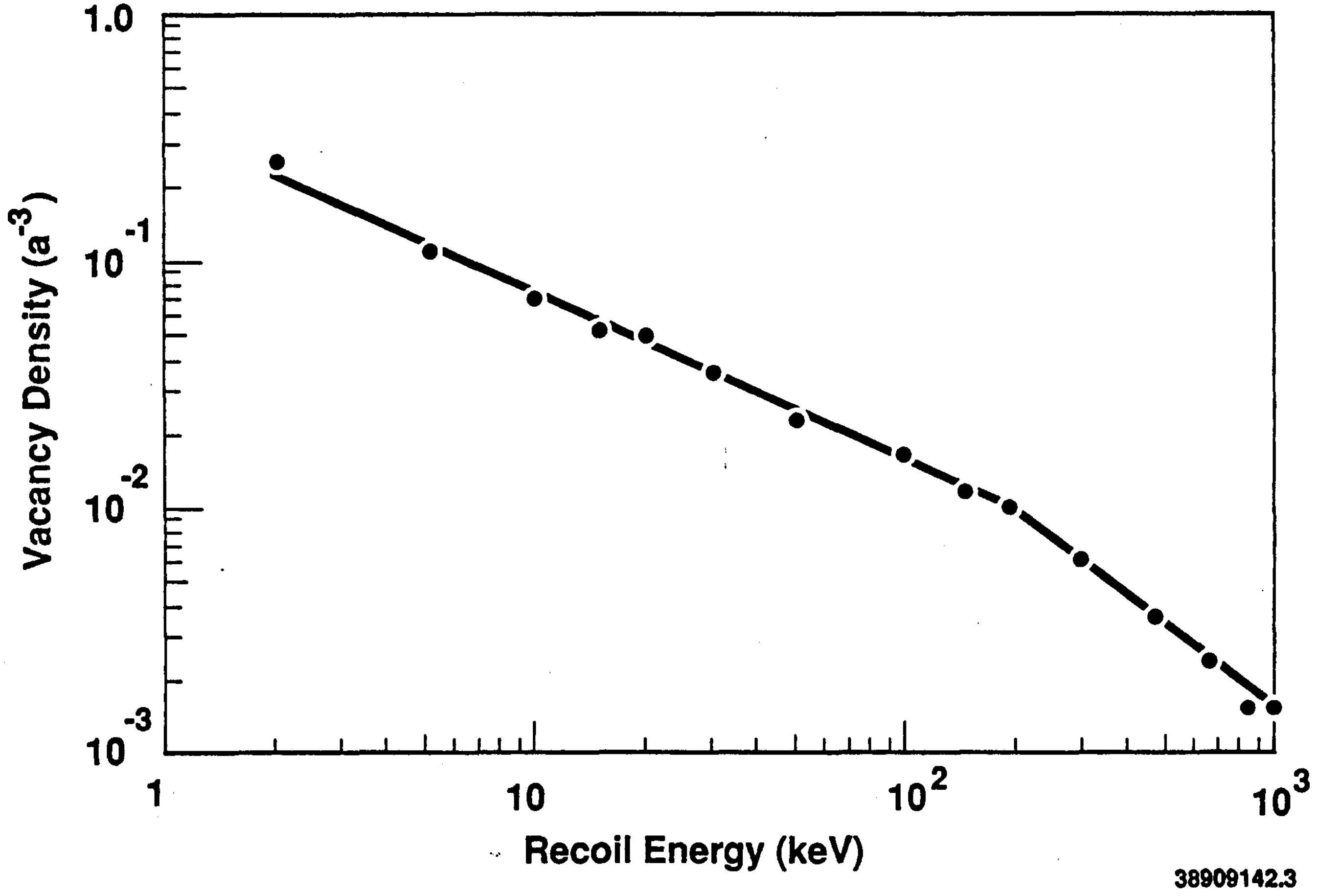

Figure 1

Average Density of Vacancies in FCC Collision Cascades

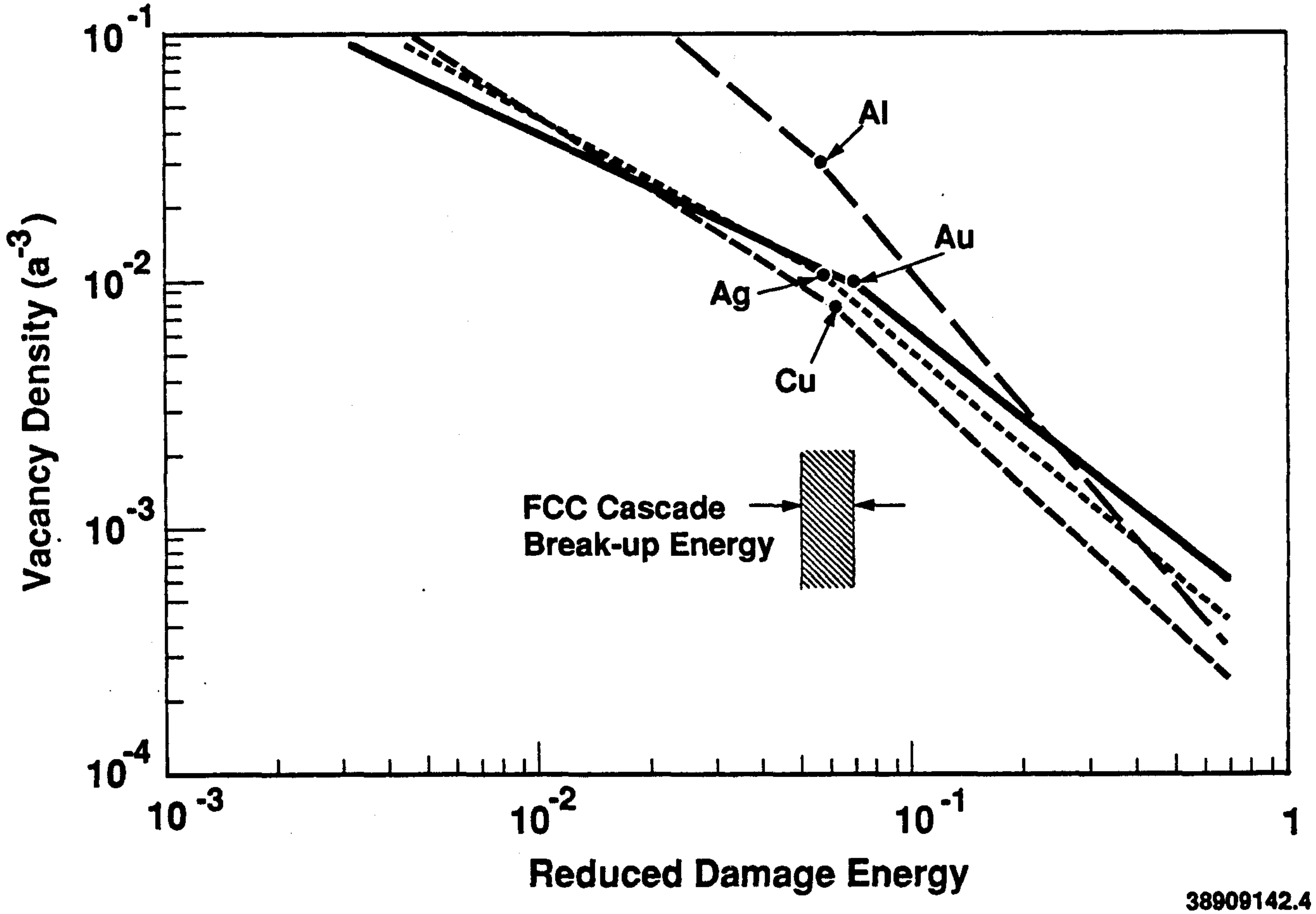

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Figure 2

Average Recoil Energies in FCC Metals

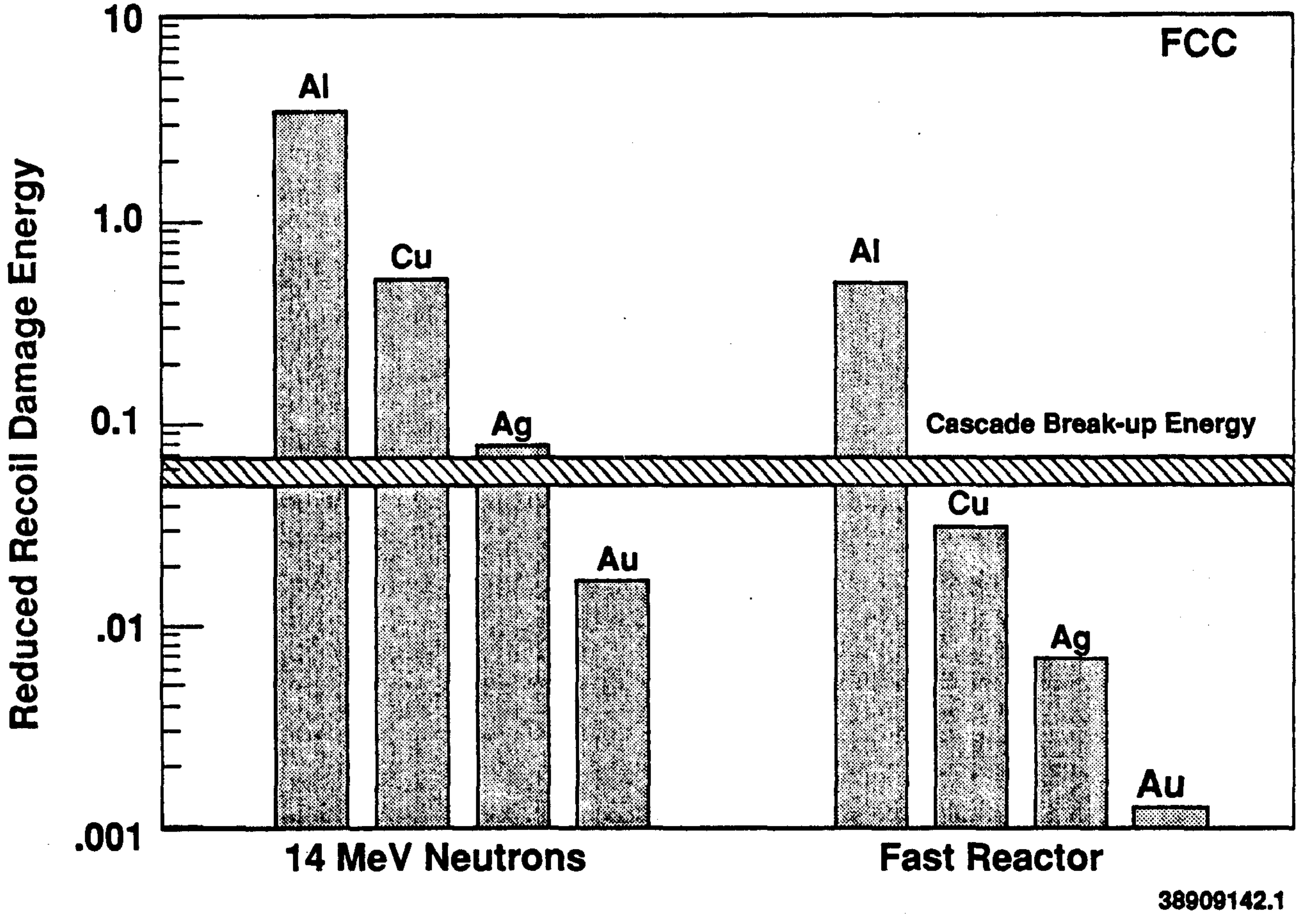

Figure 3a

Average Recoil Energies in BCC Metals

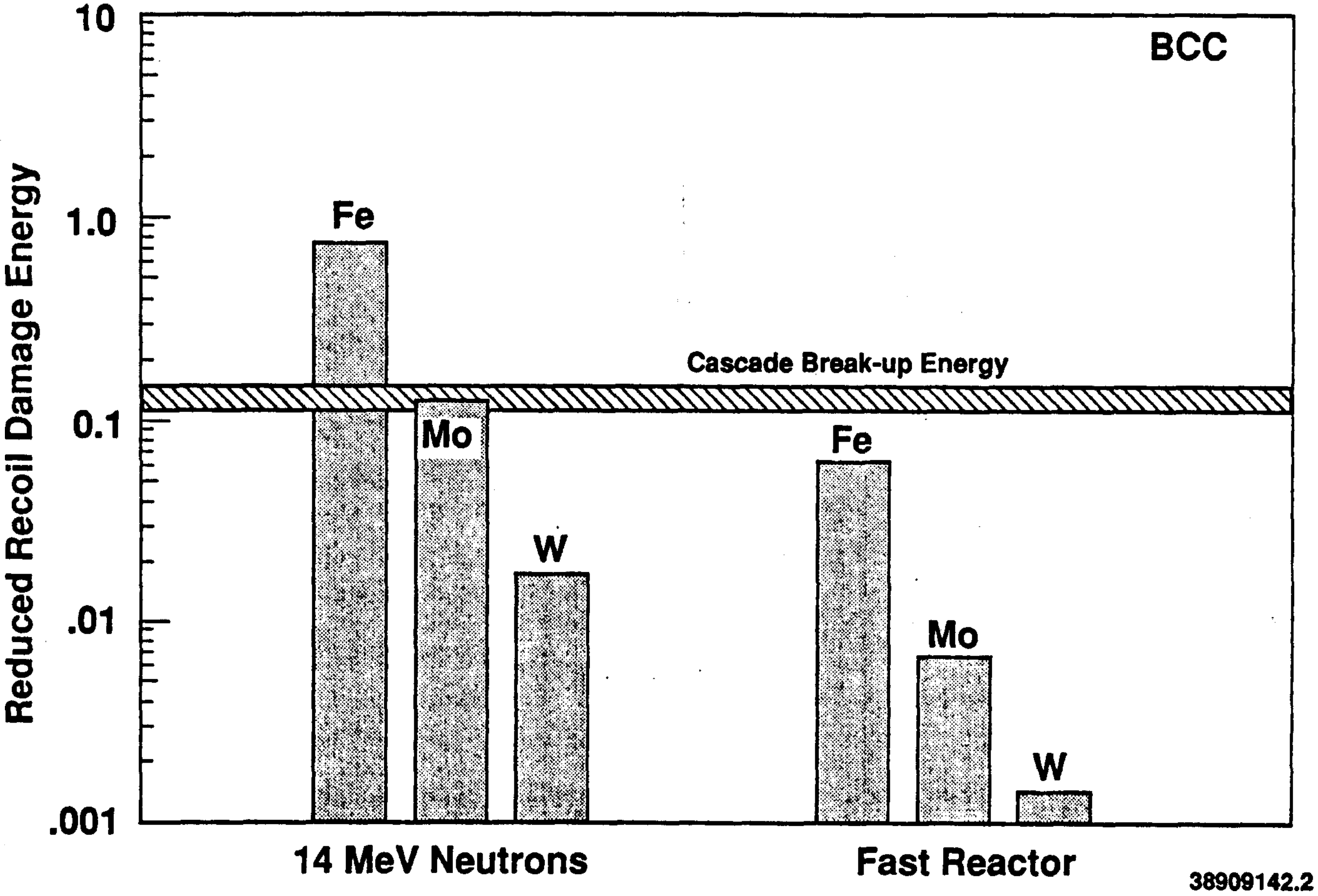

Figure 3b