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NUCLEATION OF VOIDS BY HIGH DOSE IRRADIATION

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The mechanisms of nucleation of voids in highly irradiated metals are discussed from the standpoint that the formation of dislocation ring is easier process than that of void. It was shown that the type of embryo to be grown to nucleous of void is determined at an earlier stage of clustesing, the simplest one is the tetrahedron composed of four vacancies formed by colliding of two di-vacancies. Discussions are presented for merits and demerits of other possible types of embryo including effects of impurity atoms.

1. Introduction

Since the first discovery of voids in highly neutron irradiated metals [1], increasing interests have been focused on the mechanisms of nucleation and growth of voids [2)3)], which might give a key for predictions of swelling and loss of ductility in metals to be used in fast breader reactor and fusion reactor. Comprehensive accounts were presented for mechanisms of growth of voids in a stage after the nucleation of them [4], but questions yet remain unsolved for the mechanisms of nucleation of voids.

Published theories treating the nucleation of voids may conveniently be classiffied into the two groups. The one concerns with the kinetic theory of formation of embryo of voids under circumstances of irradiation [5)6)8)]. The theories predict the critical size of nucleous of voids and its nucleation rate under various production rates of vacancies and interstitials. In these theories, however, various constants used in kinetic equations are assumed. Moreover, no disscussions were made on the possibility of two types of vacancy cluster, embryo of void and that of dislocation ring, though it is considered to be an essential problem in the formation of voids.

The other concerns with the calulation of the constants stated above based on some appropriate simulation model of real crystals [8)9)10)11)12)]. Unfortunately, however, the results of caluculations depend largely on the assumptions employed for interatomic forces, and discrepancies yet remain even in qualitative predictions for relative magnitude of formation energy of the two types of vacancy clusters.

This paper present a comment on the discussion for critical stage in formation of embrio of voids by criticizing the previous theories.

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2. The mechanism of formation of embryo of voids

According to the nucleation theory of void [5)6)7)], the time rate of change of various vacancy cluster concentrations n_i is give by

$$\frac{dn_1}{dt}=-c_1n_1+e_2n_2+c_2'n_2-c_1'n_1+p, \tag{1}$$

$$\frac{dn_i}{dt}=c_{i-1}n_{i-1}+(e_{i+1}+c'_{i+1})n_{i+1}-c_in_i-(e_i+c_i')n_i,\ i\geq 2, \tag{2}$$

where c_i; the capture rate of vacancy by cluster of i vacancies,
c_i'; the capture rate of interstitial by cluster of i vacancies,
e_i; the escape rate of vacancy from cluster of i vacancies,
p; the production rate of vacancies by irradiation.

Under the assumption of quasi-steady state, the statistical distribution of clusters n_i and the time rate of production of nucleous of void are calculated by the methods paralell to classical nucleation theory. The critical size of nucleaous, the cluster of maximum free energy, is estimated as $i\sim 10$.

In these theories however all clusters of vacancies are assumed to be embryo of void, and no distinctions were made on the differences in types of clusters. As is well known, geometrical forms of clusters are classified into the two main types [8)9)], the dislocation ring type* and void type. In almost all cases, for instance annealing of quenched metals or reactor irradiated metals and observations of metal foils irradiated by electron in high voltage electron microscope, it is well known that dislocation ring is formed as a results of coagulation of vacancies, and the formation of voids is rare cases rather limited to high dose irradiation. [13)] Then what is the distinction in mechanism, whereby the voids are nucleated. No clear explanations were presented in the previous theories.

As is well known, i vacancies clustered in an atomic plane form a dislocation ring by collapsing, which is designated by V_{id}. V_{3d} may be smallest one which have character of dislocation ring. Taking into account the nature of stress field around the clusters the interaction of a vacancy V_1 with these clusters yield the reactions,

$$V_{3d}+V_1\rightarrow V_{4d}, \tag{3}$$

$$V_{4d}+V_1\rightarrow V_{5d}, \tag{4}$$

$$\cdots\cdots\cdots\cdots.$$

On the oter hand, the smallest cluster of void type is cluster of four vacancies which have the form of tetrahedron, and is designated by V_{4v}.

* More precisely we must consider Frank sessile dislocation loop, perfect dislocation loop and stacking fault tetrahendra in FCC metals [8)9)], but first two types are not distinguishable for small clusters under consideration, and we shall not consider the case where the third type play important role for simplicity of discussions.

Cotterill and Doyama showed that the formation energy of void type is larger than that of dislocation ring type for $i=4$ in FCC metal[10)11)]. We may suppose the same for all $i>4$, and the difference increase as the number i increase. On the contrary Johnson[8)] and Beeler[12)] obtained the reverse results that the formation energy of void is smaller than that of dislocation ring for $i\lesssim 2000$.

The difference between the two results is due to the different assumptions for inter-atomic forces used in these calculations. The former used the Morse's potential for 62 neighbouring atoms in FCC lattice while the later used the Johnson's potential for 12 and 14 neibouring atoms in FCC and BCC lattices respectively. Further investigations for the cause of the difference may be important and interesting problem, but we may show here that the conclusion of later produce some difficulties and contradictions in explaining the experimental evidences.

In oder that the void be transformed to the dislocation ring by aid of thermal vibration, it must overcome the potential barrier of the disc shape cavity, so that the transformation is considered to be inhibited for clusters larger than $i\sim 6$ as seen in simple calculations. Thus the two types of the clusters, the void and the dislocation ring, are considered to be fixed in their early stage of growth.

Then if the formation energy of void is smaller than that of dislocation ring for $i\lesssim 2000$, the nucleation theory predicts that the probability of formation of voids is always larger than that of dislocation ring. This conclusion contradicts with the experimental evidences stated above.

Then we shall consider possible reactions for the formation of the embyo of void, in which we shall take into account di-vacancy V_2 together with single vacancy V_1, since V_2 move more easily than V_1. A promising one is the reaction,

$$V_2 + V_2 \rightarrow V_{4v}\ , \tag{5}$$

this accompany the alternative

$$V_2 + V_2 \rightarrow V_{4d}\ . \tag{6}$$

The choice of (5) or (6) depend on the direction of approach of the two divacancies, *i.e.* V_2 may accompany the dipolar stress field and the two parallel divacancies may attract each other in case of (6), but the interaction may not be so strong as in the case of (3). The interaction is supposed to be weak in the case of the reaction (5) too, where the two di-vacancies approach perpendicularly to each other.

When the probability of reaction (5) is much smaller than these of (3) and (6) the reactions assumed in the above equations may provide a explanations for the experimental evidences. The deduction, however, might be too strict in the meaning that the mutual transformation of the types of clusters are restricted down to smallest size. In actual cases possible reactions of the formation of embryo of voids may include the reactions,

$$V_{3d}+V_1\rightarrow V_{4v} \qquad (7)$$

$$V_{4d} \quad \rightarrow V_{4v} \qquad (8)$$

$$V_{3d}+V_2\rightarrow V_{5v} \qquad (9)$$

The reactions might not be inhibited when the temperature is high, but is considered to be smaller than (5) at low temperatures since the reactions require additional works to make bindings as explained previously.

In these cases if the formation energy of dislocation ring is smaller than that of void the nucleation theory may explain the experimental evidences.

As for the effects of impurity atoms, such as substitutional, interstitial impurities and α particles, on the nucleation of voids, the rate of formation of embryo of void may be increased through the reactions; a vacancy trapped to a impurity atom may produce immobile di-vacancy by absorbing V_1 thus increase the number of V_2 in (5), or impurity trapped V_{3d} and V_{4d} may assist the reactions (7), (9) and (8) respectively by lowering the potential barrier of each processes.

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