NUCLEATION OF VOIDS

Progress Report

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Abstract

Homogeneous nucleation theory has been generalized to include the nucleating effects of both mobile and immobile gas atoms in a material supersaturated with both vacancies and interstitials. Sample calculations were made using properties typical of those of nickel and helium. We found that at the low damage rates typically found in reactors, nucleation occurs on gas atoms, while under typical accelerator conditions, homogeneous nucleation will dominate.

At the start of the present contract period, we (Dr. Hartmut Wiedersich, Materials Science Division, Argonne National Laboratory and myself) had been able to: 1) show how to generalize nucleation theory to correctly account for the anti-nucleating effect of interstitial atoms on void nucleation, 2) make extensive sample calculations using properties typical of those of nickel, thus giving results which were useful for actual reactor materials at least semi-quantitatively¹, 3) show how to account for the nucleation effect of immobile gas molecules² (but did not have time for sample calculations).

During the present contract period, we have made extensive sample calculations³ of the nucleating effects of immobile gas using properties typical of those of nickel and helium. We found that at the low damage rates typically found in reactors, nucleation would, at normal reactor temperatures, occur primarily on vacancy clusters containing at least one gas atom, while under typical accelerator conditions homogeneous nucleation would, at reasonable gas atom concentrations, dominate. This raised some question about the extent to which results obtained in accelerator experiments could be used to predict void formation in reactors since the fundamental nucleation mechanism changes. This theory was of course limited to immobile gas, i. e., we assumed that the gas was sufficiently insoluble in the lattice that once it entered a void, the probability that it could escape again was negligible. Such an assumption is probably a very good one at typical reactor temperatures, but as the temperature increases it becomes less so.

Fortunately, the nucleation problem is also soluble in the other limit, mobile gas atoms. It was apparent to others too that such was the case, but their efforts to obtain a useful result were not successful. Loh⁴ attempted to do so by combining Reiss' binary nucleation theory⁵ with our previous work¹. However, as Russell⁶ has shown, a straightforward combination of these two theories leads to an internally inconsistent theory. Therefore, Russell⁷ attacked the problem by considering mobile gas, but only when there are no excess interstitials, a case not relevant to the nuclear reactor environment.

At the time of the previous renewal, we had an idea how this problem might

be attacked. Our idea turned out to be valid. Attached to the back of our proposal is an interim version of our paper⁸ (I delayed submitting this progress report and renewal proposal in hopes of being able to also include the final version of this paper. The paper is completed and the results obtained are correct, but the wording of sections 3.1 and 3.2 which explain our computational procedure need revision before it can be submitted for publication).

In this paper, we show that qualitatively similar results are obtained in the mobile gas limit and in the immobile gas limit. We find that at the low damage rates ($\leq 10^{-5} \text{ sec}^{-1}$), 10^{-10} atom fraction of helium can dramatically increase the void nucleation rate and lead to void formation under conditions where no voids would be observed without helium. Higher concentrations of helium are required to induce void formation under accelerator conditions, where the damage rates are greater and voids might nucleate even without helium.

These results are in qualitative agreement with experiment. Sufficient helium is present in neutron irradiated metals to promote void formation and many accelerator and electron beam experiments have shown that helium promotes void formation. However, voids have been observed in ion, proton and electron irradiated samples without helium.

These conclusions suggest that considerable care must be taken in designing accelerator experiments to simulate void formation in reactors. After verification of our predictions in accelerator experiments on helium doped materials, it should be possible to use this theoretical work to design valid reactor simulation experiments.

As of December 31, 1973, Dr. Joseph L. Katz will have devoted 96% of the time or effort called for by this contract; the 4% additional effort can be expected during January 1974.

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