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Absence of Irradiation Growth in Alpha-Uranium above 430°C

The theory of Seigle and Opinsky (*1*) explains growth of alpha-uranium on irradiation by anisotropy of diffusion of interstitial uranium atoms and of vacancies in orthorhombic lattice. To show the usefulness of and to provide a possibility of testing this theory, the

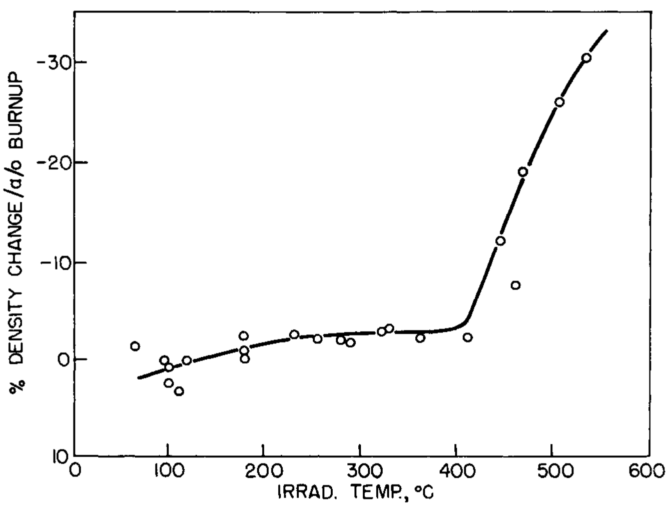

FIG. 1. Density change per atomic per cent burnup in uranium as a function of irradiation temperature after J. H. Kittel and S. H. Paine (*7*).

authors have advanced a few predictions. One of these, namely, that the irradiation growth at very low temperatures where diffusion is very slow should be low has since been confirmed by Kunz and Holden (*2*). Another prediction concerning the dependence of irradiation growth upon the crystal size of the samples, due to experimental difficulties, has been only partly confirmed by the work of Resnick and Seigle (*3*). The prediction, however, that irradiation growth should double on increasing the irradiation temperature from 200 to 500°C. apparently did not materialize. Single crystals of alpha uranium irradiated by Plail and Wyatt (*4*, *5*) at 350°C grew as expected in the [010] direction but they did not grow at all when the temperature of irradiation was increased to 450°C. This finding seemingly invalidates the anisotropic diffusion theory and also any present or future theory of irradiation growth of alpha uranium which rests upon diffusion or on differences between rates of mass transfer and of vacancy transfer (*6*).

The purpose of this note is to advance an opinion that such a conclusion need not necessarily be true and to adduce facts and arguments which permit one to consider such theories of growth as basically valid, in spite of the absence, of the growth when varying the irradiation temperature from 200 to 450°C.

The experimental facts collected by Kittel and Paine (*7*) are for the present purpose important. These are shown as Fig. 1 which illustrates how the ratio of per cent decrease in density to per cent of atom burnup changes as a function of the temperature of irradiation. It may be seen in this figure that up to an irradiation temperature of about 430°C this ratio remains approximately constant but that it rapidly decreases past this temperature. This rapid decrease of the ratio is undoubtedly caused by swelling, that is by the nucleation and growth of the Xe and Kr filled pores.

The appearance of a multitude of discrete pores within the body of irradiated alpha-uranium at 430° undoubtedly subdivides the continuity of the originally sound, void-free uranium. A multitude of internal sinks is thereby created into which the diffusing vacancies and uranium interstitials can be trapped. Under these circumstances one can suspect that the experiments of Plail and Wyatt failed to confirm the expectation of the increase of the rate of irradiation growth with the increase of the irradiation temperature because of poor choice of the test conditions. It is rather obvious now that this verification should have been conducted under adequately comparable conditions, which can only be secured by measuring the rates of irradiation growth at two or more temperatures all of which are lower than 430°C. This is essential since, with the multitude of newly created sinks, above this temperature one cannot expect any directional growth of the outside dimensions of oriented polycrystals or of single crystals. At best, as a result of the differences in the rates of diffusion of vacancies and interstitials in the different crystallographic directions one can expect some ovalization of the originally round pores. It is doubtful, however, whether such ovalization can be observed and distinguished from ovalization due to anisotropy of alpha-uranium, especially since surface diffusion and spheroidizing of the holes by creep under the influence of stresses resulting from the pressure of gas will tend to restore the spheroidicity. Thus the directional irradiation growth above 430° can be expected to degenerate into swelling which is three-dimensional. Under the circumstances it would seem to be prudent to refrain from claiming, at least for the time being, that absence of directional growth in single crystals irradiated at 450°C places diffusion based theories of irradiation growth in serious difficulties. It is felt that future experiments such as careful re-running of Plail and Wyatt's experiments at temperatures lower than 430°C are needed to decide the case.

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