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TITLE

HYPOTHESIS CONCERNING IRRADIATION EMBRITTLEMENT OF URANIUM

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HYPOTHESIS CONCERNING IRRADIATION EMBRITTLEMENT OF URANIUM

by

E. C. Wood

SPECIAL RE-REVIEW
FINAL DETERMINATION

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BY JP Denoux DATE 6-26-81BY JW Jordan DATE 6-30-81RESTRICTED DATA

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Introduction

In discussions with a number of people at H.A.P.O., K.A.P.L. and A.N.L. a hypothesis has been evolved which appears to fit available information concerning irradiation embrittlement of uranium as well as indicate a possible solution to the problem. The purpose of this memorandum is to expound this hypothesis as an aid to those working with the problem. Since it embodies the ideas of many people, no claim to unique authorship is implied.

Discussion

Recently M. B. Reynolds at K.A.P.L. in studying evolution of radioactive krypton from small irradiated specimens of uranium observed that the total gas evolution from the sample was markedly increased by cycling the sample through the alpha, beta, and gamma phases. The rate of gas evolution, initially high on heating, dropped off rapidly until the sample had again been cooled into the alpha phase and was re-heated. A like sample held in the gamma phase for several days evolved gas very slowly until after several days it all came off at once. Plots of the total krypton evolution versus time at temperature for the two samples are shown in KAPL-1039, and appear somewhat as shown in Figure 1.

Reynolds in explaining these data suggests that the gas appears slowly at the grain boundary gradually filling up vacancies until such time as the grain boundaries are filled. After this, on further holding, it ruptures out of the sample. In explaining the effect of cycling he notes that one of the effects of such thermal cycling through phase changes is to shift the position of the grain boundaries and consequently to make available more total grain boundary volume for absorption and release of the gas.

This experiment suggests, in agreement with intuitive ideas, that the rate of diffusion of fission products from the grain to the grain boundary is very low and is, in fact, probably the same as the rate of self diffusion of uranium. The grain boundary, having many more dislocations than the grain, will act as a sink--absorbing fission product atoms until the vacancies are filled. The experience of Reynolds in obtaining greater gas evolution from the sample on changing the location of the grain boundaries also suggests that the diffusion rate through the grain boundary material is much higher than the diffusion rate from the grain to the grain boundary.

This explanation suggests that on irradiation fission products formed in the grain will then move slowly toward the grain boundary, forming a concentration gradient from the interior of the grain to the boundary which, because of the essentially uniform rate of formation within the grain, will be a parabolic function of distance from the boundary. The rate of ascent of the concentration function will be proportional to the power level when the condition is reached at which the rate of diffusion from the grain to the boundary equals the rate of formation. Thus as shown in Figure 2 a large grain will have a high concentration of fission products at its center, while a small grain would be expected to have a much smaller concentration.

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Total Kr Evolved

Days

Figure 1

Approximate Fission Gas Evolution (After Reynolds)

- a. Cycled between ≈ 900 C and room temperature
- b. Held at ≈ 900 C

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Fission Product Concentration

Grain Boundary

Small Grain
Boundary

Large Grain
Boundary

Figure 2

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To explain the embrittling effects it is necessary to postulate that the effect of fission products in the grain boundary is less severe than the effect of fission products in the grain. This is generally indicated by the work done with the hardness surveys of irradiated material at Hanford and by the studies of ductility of irradiated samples at K.A.P.L. In all of this work it was shown that the effect of irradiation in damaging the structural properties was a minimum at the center of the slug where the higher temperatures would promote rapid diffusion toward the grain boundary. One is thus lead to hypothesize that uranium having fine grain structure will suffer less embrittlement on irradiation than will uranium with a coarse structure. This is confirmed to some extent by experience at Argonne National Laboratory with irradiated samples of unalloyed and uranium 2 a/o zirconium alloy. The latter alloy was made in order to obtain fine grain size in a cast structure so that the resulting sample would be both fine grained and randomly oriented. This alloy was found to be stable although embrittled under irradiation up to 2 percent burnup--equivalent to about 20,000 MWD/T--while the unalloyed uranium was found to be exceedingly brittle and friable at lower burnups.

No direct comparison between this experience and Hanford experience is possible because of the highly different condition of irradiation, nor is it certain that the improvement noted results only from the fine grain size and not from some other effect of the alloy addition. However some other confirming data may be available when similar powder metal samples which are being irradiated have been examined.

Certain experiments are suggested by this hypothesis: first, it is suggested that the cored bonded slug be tapped and the concentration of fission gas in the core be determined, second, it is suggested, as is already planned, that the fine grain powder in metal compacts now being irradiated be irradiated to as high a level as possible in order that the embrittlement of these at maximum irradiation may be compared with present standard material.

With respect to unbonded slugs Lang has suggested a mechanism of failure attributed to the release of fission gases within the small volume between the can and the slug. This mechanism also suggests a possible solution which may have been inadvertently used by the British. Zinn in his report on a visit to the British Atomic Energy Establishment notes that the helium with which they are known to fill their unbonded slugs may be introduced from a capsule containing helium under pressure which is inserted into the slug before canning and ruptured after seal is made. The volume of such a capsule would provide a convenient place for fission product gases to collect during irradiation and would minimize the pressure of fission product gas achieved during irradiation inside the can. This suggests that an unbonded slug having a relatively large space for gas to collect should not rupture by the mechanism which Lang postulates and that this

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-6-

HW-36111

mechanism might be avoided on solid unbonded slugs by providing in the cap or base a void volume large enough to minimize the pressure.

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