LBL-13205 c.2

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

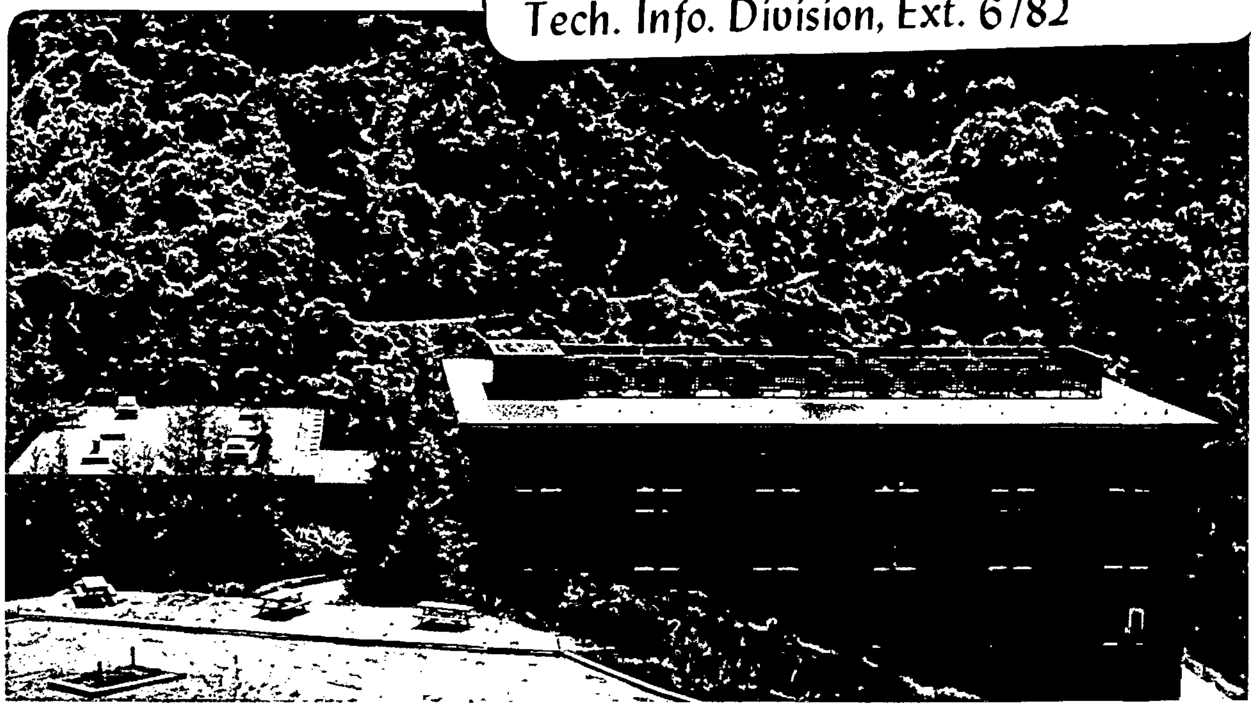
Materials & Molecular Research Division

To be presented at the 1981 Winter Meeting of the American Nuclear Society, San Francisco, CA, November 29 - December 4, 1981

THE EFFECTS OF LONG-TERM EXPOSURE TO SIMULATED FISSION PRODUCTS ON THE PROPERTIES OF ZIRCALOY-2

Rajiv Kohli

August 1981



Prepared for the U.S. Department of Energy under Contract W-7405-ENG-48

DISCLAIMER

LBL-13205

THE EFFECTS OF LONG-TERM EXPOSURE TO SIMULATED FISSION PRODUCTS ON THE PROPERTIES OF ZIRCALOY-2*

Rajiv Kohli

Materials and Molecular Research
Division of the Lawrence Berkeley
Laboratory and the
Department of Nuclear Engineering
University of California Berkeley
Berkeley, California 94720

Paper for Presentation at the 1981 Winter Meeting of the
American Nuclear Society, November 29-December 4, 1981
San Francisco, California

*Work performed in conjunction with Österreichisches Forschungszentrum Seibersdorf, Austria

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U. S. Department of Energy under contract # W-7405-ENG-48.

THE EFFECTS OF LONG-TERM EXPOSURE TO SIMULATED FISSION PRODUCTS ON THE PROPERTIES OF ZIRCALOY-2

Extended Abstract

Although considerable research has been carried out on the Pellet-Clad-Interaction (PCI) failure problem in LWR fuel rods, the nature of the chemical environment involved in such failures of the zircaloy cladding is still not definitely known. Further, the fuel rods are operated for long times during which they are constantly exposed to the fission product chemical environment. A comprehensive research program was, therefore, initiated to obtain useful information on the effect of prolonged exposure of Zircaloy-2 to fission product environments. Some results from the ongoing program have been reported previously [1,2]. This paper will present some further results which, we believe, are of major significance and have a direct bearing on the PCI failure of zircaloy-clad fuel rods.

First, a detailed thermochemical assessment was made of the chemical states and the transport properties of the fission products in LWR fuel rods. In addition, a series of compatibility tests with a variety of simulated fission elements and compounds was performed to characterize the reaction behavior of Zircaloy-2. Based on this information various simulated fission products were tested for activity in the embrittlement of Zircaloy-2. Room temperature tensile tests were carried out on flat specimens after exposure to high purity argon and to the individual products, I + Se, I + Te, Ag, Nb, Ru, Mo, W, BaO, SrO, Y_2O_3, CeO_2,

La_2O_3, and Gd_2O_3, from 50-8000 hours in the temperature range 350-700 ^{o}C. This choice of experimental conditions was dictated by a) the need to simulate normal fuel rod operating conditions (∿350 ^{o}C, >8000 hours); and b) the need to consider conditions prevailing in "hot spot" regions (>500 ^{o}C, long times). In addition, we hoped that annealing at higher temperatures for short times might provide a limited basis for screening the more aggressive fission products for lower temperature annealing.

Annealing in the temperature range 350-500 ^{o}C resulted in a gradual but definite decrease in the room temperature ductility of the Zircaloy-2 specimens with annealing time (see Fig. 1). The specimens broke with high reduction in area and exhibited only ductile fracture morphology, except for specimens annealed in the rare earth and the alkaline earth oxides. These specimens were severely embrittled, although annealing times longer than 1500 hours were required for embrittlement (Fig. 1). By contrast, the specimens annealed in these oxides at 700 ^{o}C were completely embrittled after only 50 hours (see Fig. 2). This can be explained since the rate of diffusion of oxygen in the metal is much slower at 350 ^{o}C, whereas at 700 ^{o}C the rate is rapid enough to saturate the thin zircaloy specimens within 50 hours. The source of oxygen here is a nonstoichiometric double oxide layer that forms on the metal surface.

Besides the oxides, the other fission products tested also showed some remarkable effects on annealing the specimens at 700 ^{o}C (see Fig. 2). Mo and W are severely embrittling, as is Ru (a typical fracture surface is shown in Fig. 3). This is primarily due to the formation of $ZrMo_2$, ZrW_2, and ZrRu intermetallics along the grain boundaries, as indicated by x-ray diffraction, Auger electron spectroscopy, and electron microprobe analysis. In the case of annealing in Ag and Nb, intermetallic compound formation causes a marked reduction in ductility of the specimens (see Fig. 2).

The implications of these results to the PCI failure problem in LWR fuel rods will be discussed in detail.

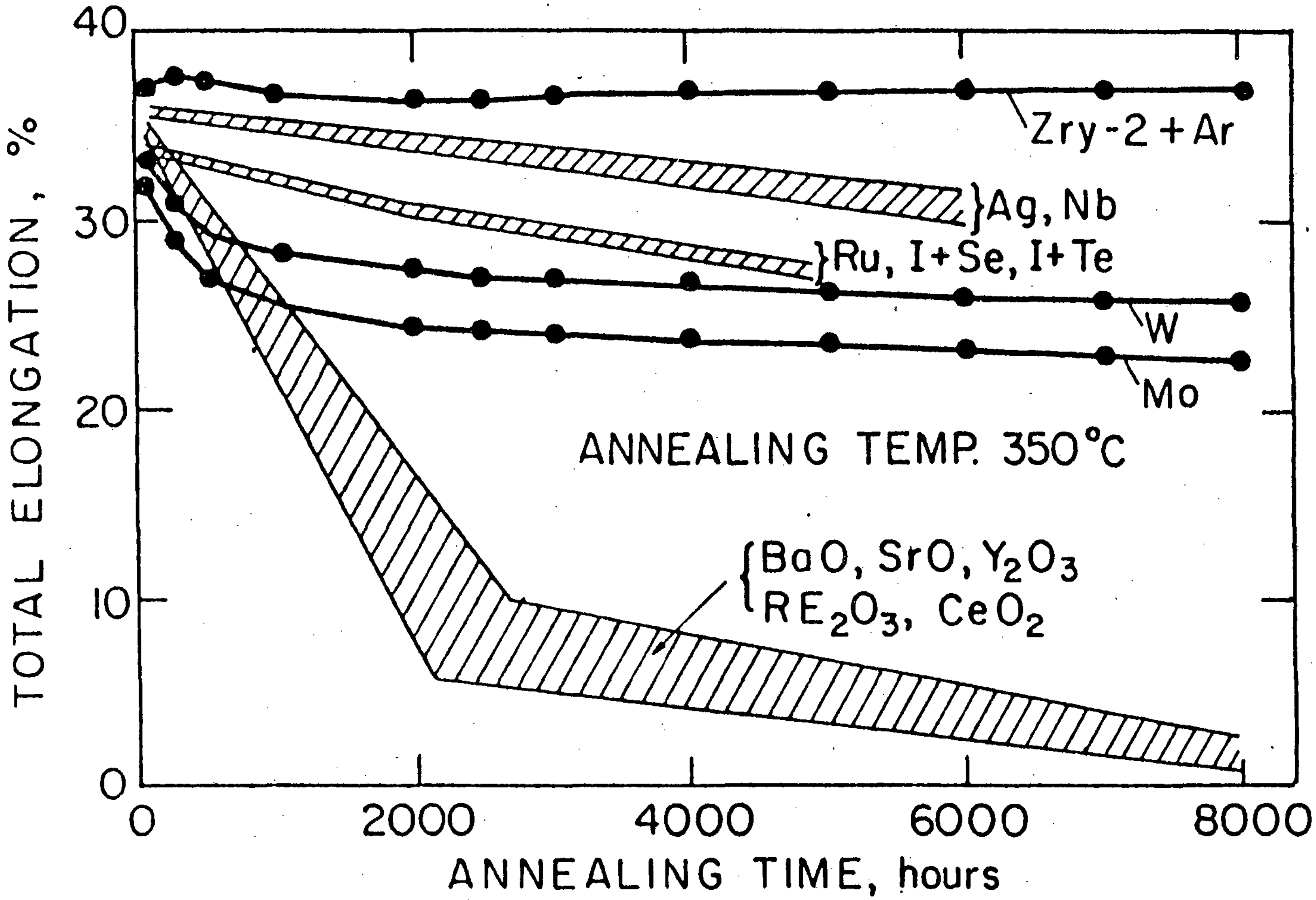
<u>References</u>

1. R. Kohli and F. Holub, Nucl. Technol., 48, 70 (1980).

2. R. Kohli and F. Holub, J. Nucl. Mater., 98, 116 (1981).

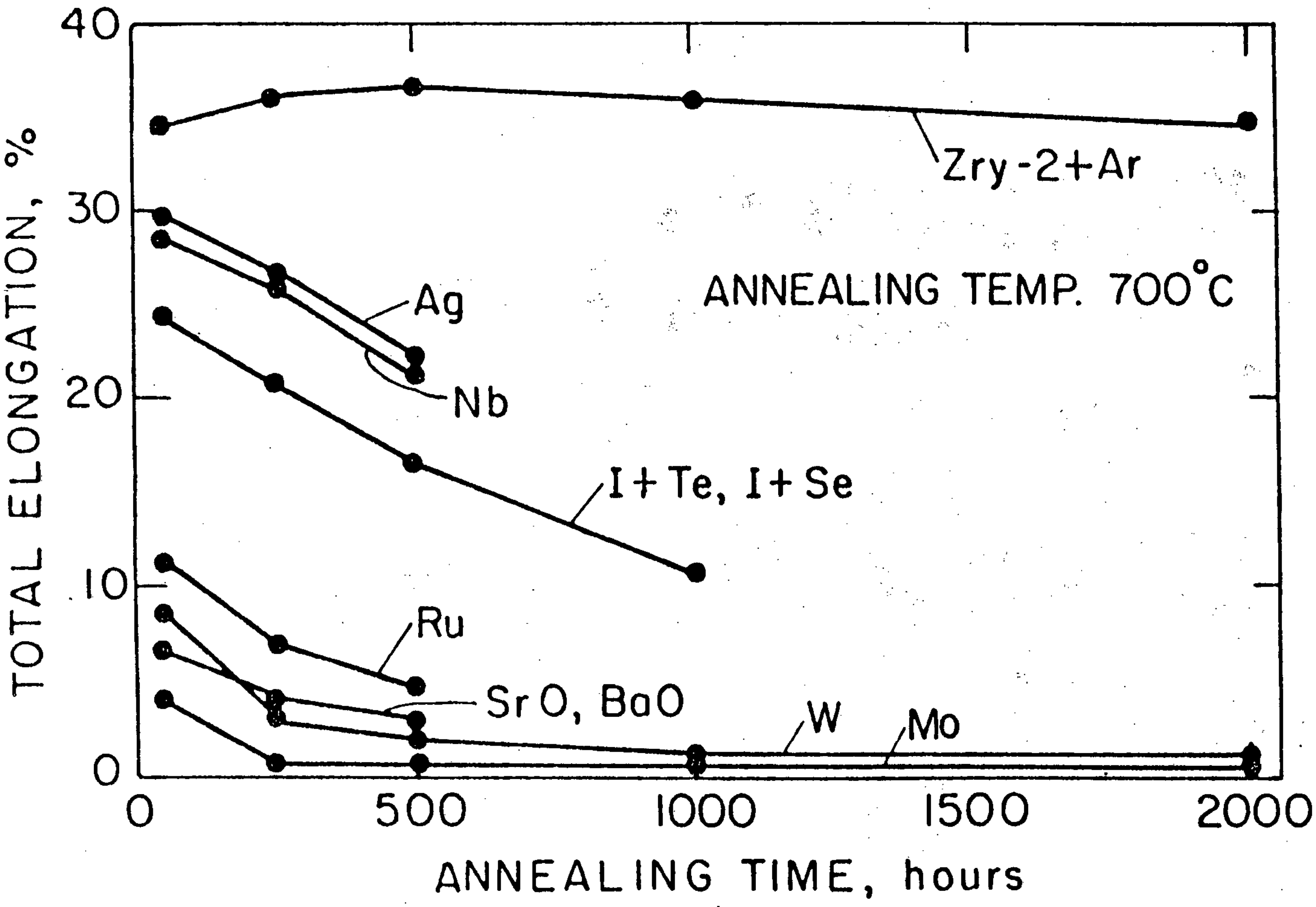
<u>Acknowledgement</u>

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U.S. Department of Energy under Contract #W-7405-ENG-48.



XBL 816-5857

Fig. 1. Variation of Total Elongation as a Function of Annealing Time for Zircaloy-2 Specimens Exposed to Argon and Fission Products at 350 °C.



XBL 816-5856

Fig. 2. Variation of Total Elongation as a Function of Annealing Time for Zircaloy-2 Specimens Exposed to Argon and Fission Products at 700 °C.

7 μm

Fig. 3. XBB816-4964

Scanning Electron Micrograph of the Fracture Surface of the Zircaloy-2 Specimen Annealed in Ruthenium at 700 °C for 250 Hours.

TECHNICAL INFORMATION DEPARTMENT
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720