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D. O. Hobson

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ABSTRACT

This study is directed toward the determination of the effects of annealing cycles with rapid heating rates, short hold times at specific temperatures, and rapid cool-down rates on the hardness of Zircaloy fuel cladding. These rapid annealing cycles are designed to provide preliminary annealing behavior data on Loss-of-Fluid-Test Reactor cladding samples. Information has been obtained on

1. the time dependence of the hardness as a function of annealing temperature, and
2. a correlation of single- and multitransient annealing relationships.

Both single- and triple-cycle transients were used; four hold times at each of five maximum temperatures comprised the data set (each portion of the triple-cycle experiments had isothermal hold times equal to one-third of their analogous single-cycle times).

It was found that there was little difference in the hardness response between single- and triple-cycle transients for a given total hold time at a particular temperature. Test temperatures range from 1000 to 1400°F (538 to 760°C) and hold times from 5 to 135 sec. The 1100°F (593°C) level was found to be the transition level for hardness changes, with shorter times (5 and 15 sec) effecting little or no hardness decrease and the longer times (45 and 135 sec) producing partially and fully annealed material, respectively. Temperatures equal to or greater than 1300°F (704°C) resulted in fully annealed material for all hold times. The 1000°F (538°C) tests produced no measurable softening.

INTRODUCTION

The Loss-of-Fluid-Test Reactor (LOFT) is to be operated by Aerojet Nuclear Company (ANC) to determine, on a large scale, the effects of various simulated reactor accident situations. Since the fuel elements of the LOFT reactor will undergo the thermal cycles involved in the various test programs, it is necessary to study their

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response to those cycles. The present study was designed to characterize, in a simple way, the effects of rapid thermal cycles on the hardness of LOFT cladding. The hardness test, specifically the diamond-pyramid microhardness test, is a sensitive indicator of the metallurgical state of the cladding, and it can determine the transition from the as-received, stress-relieved state of the cladding to the fully annealed condition.

This study was designed to provide information on two aspects of the thermal response of LOFT cladding. The first involves the metallurgical response of the material to very short elevated-temperature exposures, which are preceded by rapid heat up and followed by a rapid cool down. What times at a specified temperature are sufficient to soften the Zircaloy cladding, i.e., produce the annealed state? The second aspect is concerned with the equivalency of temperature cycles. Do three cycles, each run for a time, t , at temperature, produce the same softening as a single-cycle run for equivalent time, $3t$, at temperature?

The present study does not address a comprehensive range of the mechanical properties variables that respond to heat treatment; such variables as yield and ultimate strengths, elongation, reduction in area, etc. Rather, it examines hardness only; but hardness is a valid indication in Zircaloy, as in other materials, of the effects of annealing and recrystallization.

EXPERIMENTAL DETAILS

The tests in this study were performed on LOFT fuel cladding; Zircaloy-4, with measured dimensions of 0.424-in. (10.8-mm) OD and 0.025 to 0.026-in. (0.63 to 0.66-mm) wall thickness. Ring specimens were cut with a lathe to 1/4-in. (6.3-mm) lengths. The rings were then instrumented with Chromel-Alumel thermocouples spot-welded directly to the ring surfaces - one lead on the outer surface and one on the inner surface at the same axial and circumferential position.

The instrumented ring was positioned for thermal cycling by a 0.24-in. (6-mm) OD quartz tube. The thermocouple leads, insulated with "two-hole" ceramic tubing, ran through the quartz and up through a slot

in the quartz tube to their attachment with the Zircaloy. The other end of the quartz tube was clamped to a sliding block that was positioned, at the inner range of its travel, by a micrometer stop adjustment. Figure 1 illustrates the equipment setup. Heat for the thermal cycle was supplied by a Model 4085 ellipsoidal, radiant-heating furnace powered by a 750-W quartz-halogen lamp and manufactured by R-I Controls, a division of Research, Inc. The lamp was driven by manual control with a 10-A Variac. The furnace is designed to reflect, somewhat diffusely, the filament of the lamp, located at one focus of the ellipsoid, into a small volume located at the opposite focus. The longitudinal position of the hot spot was determined by a temperature scan along the axis of the furnace at a constant, low power level. This position was fixed by locking the micrometer stop to limit the inward travel of the specimen.

Specimen temperature was monitored with a two-pen Hewlett-Packard chart recorder driven at 0.2 in./sec (5 mm/sec) chart speed. One pen recorded specimen temperature and the other drew a constant-temperature line at the required maximum test temperature and served as the reference line for manual control.

Specimen cool down was achieved by withdrawing the specimen holder assembly from the furnace so that the specimen ring entered a stream of flowing argon gas. The furnace power was simultaneously turned off.

Three types of thermal cycling tests were used in this study. The first was a single-cycle transient that involved a rapid, nominally 200°F/sec (111°C/sec), heat-up rate from 600°F (316°C) to the maximum temperature and a rapid cool down to below 600°F. The second involved three successive transients with heat-up and cool-down rates similar to the single-cycle transient, but with the cool down after the first two cycles arrested as the temperature dropped below 600°F and reheat started. The specimen was cooled to room temperature after the third cycle. The total time at the maximum temperature for a particular triple-cycle specimen was made equal to the total time at maximum temperature for an analogous single-cycle specimen.

Fig. 1. Schematic Diagram of the Transient Annealing Apparatus.

The third type of test was essentially a "thermal spike" test in which a ring specimen was rapidly heated to a preselected temperature and immediately cooled from that temperature. Figure 2 illustrates the six test transients that were obtained.

Cycled specimens were mounted in epoxy, wet ground on 180- through 600-grit polishing paper and chemically polished with a 46:46:8 $H_2O:HNO_3:HF$ solution to remove cold work and provide an appropriate surface for the hardness impressions.

Hardness testing was performed with a diamond-pyramid indenter with a 1-kg load. Initially, four midwall readings were taken from each ring, with the impressions spaced every 90° from the thermocouple attachment position. It was found later that the short-time tests produced specimens with varying hardnesses in the circumferential direction due to uneven heating rates that created temperature differences around the specimen. Hardness readings for these specimens were repeated and concentrated near the thermocouple position - the position of maximum confidence in temperature.

Fig. 2. Transient Heating/Cooling Cycles with Zero Hold Times.

In order to determine the extent of circumferential differences in heating rate and thermal behavior within a single specimen, a ring was equipped with two thermocouples, each with its leads attached to the inner and outer surfaces of the ring as before. One thermocouple held the ring in approximately the same spatial position occupied by the previous test rings, and the other thermocouple was attached 180° around the ring. The ring was not in contact with the quartz tube, but was held by the four thermocouple wires. Figure 3 illustrates the thermocouple/ring geometry and shows the temperature-time relationships through two heating cycles. The solid curve represents the "control" thermocouple, the output of which was monitored to determine the manual setting of the power level to the furnace. The first curve was obtained from an arbitrary cycle to approximately 1100°F (593°C) with a 5-sec hold time. The maximum difference between the two sides of the ring was approximately 70°F (22°C), with the auxiliary thermocouple exhibiting a faster thermal response on both heating and cooling. The same specimen was then cycled with more careful control for 45 sec at 1100°F (593°C). Again, the auxiliary thermocouple showed a more rapid thermal response under transient conditions. However, under steady-state conditions, the auxiliary thermocouple showed a steady decrease in temperature relative to the "control" thermocouple.

The cause of this behavior is unknown, but a possible reason is a shifting of the hot zone of the furnace with time at temperature. No measurements of radial temperature variation were made and it is possible that the slightly off-center position of the specimen could have put it in a thermal-gradient region. The hot-zone shift could be occasioned by movement of the lamp filament with time.

RESULTS

The results of this study demonstrate that significant annealing of the Zircaloy tubes can occur in relatively short times at temperatures of 1200°F (649°C) and above. Table 1 lists the hardnesses that were obtained with the tabulated test conditions and Fig. 4 shows the hardness results as a function of maximum nominal test temperature.

Fig. 3. Temperature-Transient Curves for Tests Made to Investigate Nonuniform Heating Behavior of Specimens.

Table 1. Test Conditions and Results of Transient Tests

Test Number	Nominal Maximum Temperature		Nominal Time at Temp. Per Cycle (sec)	Heating Rate		Circumferential Hardness, DPH				Remarks
	(°F)	(°C)		(°F/sec)	(°C/sec)	At 90° Intervals		Average		
<u>Single-Cycle Transients</u>										
A-1	1100	593	5	178	99				237	Averaged near TC position
A-2	1100	593	15	143	79				230	Averaged near TC position
A-3	1100	593	45	156	87				218	Averaged near TC position
A-4	1100	593	135	185	103	180	182	180	184	Averaged near TC position
B-5	1200	649	5	200	111	185	184	181	177	Overhot Temp significantly
B-6	1200	649	5	200	111	193	196	193	192	1st repeat of B-5
B-7	1200	649	5	200	111	192	187	199	203	2nd repeat of B-5
B-8	1200	649	15	168	104	181	184	185	177	
B-9	1200	649	45	214	119	187	185	185	185	
B-10	1200	649	135	188	104	186	187	181	185	
C-9	1300	704	5	219	122	179	179	179	181	
C-10	1300	704	15	260	144	185	183	184	180	
C-11	1300	704	45	250	139	182	176	178	181	
C-12	1300	704	135	279	150	184	184	184	187	
D-13	1400	760	5	276	153	183	180	179	184	
D-14	1400	760	15	370	178	185	177	180	179	
D-15	1400	760	45	250	139	178	184	183	175	
D-16	1400	760	135	348	193	182	182	182	178	
<u>Triple-Cycle Transients</u>										
E-17	1100	593	5	143, 227, 280	79, 126, 158				210	Averaged near TC position
E-18	1100	593	15	156, 190, 250	87, 58, 139				207	Averaged near TC position
E-19	1100	593	45	172, 200, 263	96, 111, 166	192	190	187	192	
F-20	1200	649	5	200, 222, 214	111, 121, 119				211	Averaged near TC position
F-21	1200	649	15	200, 176, 276	111, 98, 98	185	182	182	177	
F-22	1200	649	45	240, 200, 240	111, 111, 131	178	187	188	182	
G-23	1300	704	5	233, 292, 280	129, 162, 156	180	175	183	181	
G-24	1300	704	15	280, 233, 233	156, 129, 139	182	178	177	182	
G-25	1300	704	45	318, 259, 358	177, 144, 204	185	175	171	182	
H-26	1400	760	5	308, 276, 233	171, 153, 131	186	177	180	180	
H-27	1400	760	15	178, 160, 242	99, 89, 134	184	177	178	187	
H-28	1400	760	45	400, 270, 267	222, 267, 148	178	192	182	182	
<u>Single-Cycle Transients</u>										
I-30	1000	538	5	168	92	238	233	237	232	
I-31	1000	538	15	108	66	239	233	224	222	
I-32	1000	538	45	125	69	239	233	232	232	
I-33	1000	538	135	160	89	234	224	229	231	
<u>Triple-Cycle Transients</u>										
J-34	1000	538	5	148, 235, 211	82, 131, 117	235	234	235	236	
J-35	1000	538	15	118, 174, 182	66, 97, 191	231	232	233	231	
J-36	1000	538	45	125, 190, 231	69, 108, 117	233	227	232	229	
<u>Single-Cycle Transients</u>										
K-37	1000	538		167	93	230	232	233	221	
K-38	1100	593		263	146	234	209	230	229	
K-39	1200	649		261	145	239	238	235	223	
K-40	1300	704		269	149	232	232	234	230	
K-41	1400	760		296	164	230	224	204	228	
K-42	1500	816		270	150	184	167	171	181	

Fig. 4. Hardness Results for all Transient Tests in This Study.

The average hardness of the as-received material was measured to be 237 DPH. All tests conducted at a 1000°F (533°C) maximum temperature were found to have average hardnesses only slightly less than the as-received material (230–235 DPH). The 1100°F (593°C) maximum-temperature tests clearly showed typical annealing behavior with the shorter test times producing little hardness drop (230–237 DPH) and the longer times producing almost fully softened material (132–192 DPH). Tests cycled to maximum temperatures of 1300 and 1400°F (704 and 760°C) were completely softened even after the minimum test times.

As seen in Fig. 4, there is no obvious trend in the annealing behavior of the LOFT cladding when subjected to either the single or the triple transient with equal total time at any particular maximum temperature. It is felt that this resulted from the very rapid heat-up and cool-down rates employed in these tests. If slower rates had been

used, it is probably that the integrated time-temperature effect would have been more pronounced and that a triple-cycle transient would have resulted in a lower hardness than would the equivalent single cycle.

In addition to those tests in which specimens were held for specific times at maximum temperature, an additional set was run with zero hold time (or as close an approximation as possible). The specimen was heated at a fast rate from 600°F (316°C) to the maximum temperature and, when it reached that temperature, was rapidly withdrawn into the gas stream for cooling. This resulted in a fast thermal spike. For instance, the 1400°F (760°C) test was heated at approximately 289°F/sec (161°C/sec) to 1410°F (766°C) and cooled at an average rate of 350°F/sec (194°C/sec) from 1410 to 1200°F (766 to 649°C) and at a gradually decreasing average rate of 160°F/sec (89°C/sec) from 1200 to 600°F. The entire test took 7 1/4 sec. The test series was run to maximum test temperatures of 1000 to 1465°F (538 to 796°C). Figure 4 shows apparent inconsistencies in the results, with the 1300 and 1465°F (704 and 796°C) specimens annealed and with the 1400°F (760°C) specimen only slightly softer than the as-received tubing. It is believed that such variations resulted from slight differences in heating and/or cooling rates in temperature ranges where very rapid annealing is possible. No effort was made to ensure that the thermocouples were attached to the tube specimens at positions of equal wall thickness from specimen to specimen, nor was special effort made to position specimens in identical attitudes from test to test. Therefore, slight differences in heating rate could have existed undetected and could have exerted an influence on the material behavior. Although the chart recorder had rapid pen travel, it was still an electromechanical device and could have missed some time-temperature-related detail near the peak turnaround temperature. It is apparent that the annealing response of the Zircaloy-4 is quite a sensitive function of the experimental thermal "spike" conditions.

Finally, a discussion of heating rate is in order. The specimens in the tests were heated at a nominal rate of 200°F/sec (111°C/sec). It is clear that manual control of furnace temperature is not the optimum method for effecting a constant heating rate from test to test. Rough

estimates of the heating rates are listed in Table 1 and vary from 100 to 470°F/sec (56 to 261°C/sec). [The average rate for all cycles was 224°F/sec (124°C/sec).] The heating rates utilized in this study were sufficiently high to cause no apparent effect on the annealing behavior. Several of the lower rates occurred in the low-temperature, short-time transients and, as seen in Fig. 4, did not produce inconsistent results. For instance, the hardness of the 15-sec 1100°F (593°C) test (A-2), with a nominal rate of 143°F/sec (79°C/sec), is not different from the 3 × 5 sec 1100°F (593°C) test (E-17), with nominal rates of 143, 227, and 230°F/sec (79, 126, and 128°C/sec). It appears that some scatter in hardness values has resulted from the different heating rates as well as the temperature overshoot in some tests. However, closer control of the thermal conditions in these tests would not be expected to change the overall hardness trends discussed above.

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