JOURNAL OF NUCLEAR MATERIALS 26 (1968) 51–76. © NORTH-HOLLAND PUBLISHING CO., AMSTERDAM

UNIAXIAL IN-REACTOR CREEP OF ZIRCONIUM ALLOYS

V. FIDLERIS

Chalk River Nuclear Laboratories, Atomic Energy of Canada Ltd., Chalk River, Ontario, Canada

Received 18 October 1967

The report summarises the experimental results of 24 uniaxial in-reactor creep tests on various zirconium alloys, covering a range of stresses from 7 to 42 kg/mm² and temperatures between 200 and 400 °C. At 300 °C increases in creep rate of up to an order of magnitude have been measured in a fast neutron flux of $0.4–1.2 \times 10^{13}$ n/cm²·sec ($E > 1$ MeV) with specimens of cold-worked Zircaloy-2. The flux induced increases were smaller with cold-worked and heat treated Zr-2.5 wt % Nb and not measurable in annealed zirconium. Prior irradiation had no significant effect on creep of Zircaloy-2 and Zr-2.5 wt % Nb in the 10^{-7}/h range, but significantly reduced the creep rate in the 10^{-5}/h and higher ranges. The stress dependence of the in-reactor creep rate appears to follow a power law in cold-worked Zircaloy-2 and Zr-2.5 wt % Nb, with a stress exponent of about 3. The temperature dependence of the in-reactor creep rate changes around 350 °C in both cold-worked Zircaloy-2 and heat treated Zr-2.5 wt % Nb. Above 350 °C the dependence can be expressed by an apparent activation energy of 50–60 000 cal/mol, while below 350 °C the activation energy value drops to about 20 000 cal/mol.

Ce travail résume les résultats expérimentaux concernant 24 essais de fluage uniaxial en réacteur sur différents alliages de zirconium couvrant un intervalle de tensions de 7 à 42 kg/mm² et de températures comprises entre 200 à 400 °C. A 300 °C, des accroissements de la vitesse de fluage atteignant un ordre de grandeur ont été mesurés dans un flux de neutrons rapides de $0{,}4–1{,}2 \times 10^{13}$ n/cm² sec. ($E > 1$ MeV) avec des échantillons de Zircaloy-2 écrouis à froid. Les augmentations induites par le flux étaient plus petites avec l'alliage Zr Nb à 2,5% en poids écroui et traité thermiquement et non mesurables dans le zirconium recuit. Une irradiation antérieure n'a pas d'effet significatif sur le fluage du Zircaloy-2 et de l'alliage Zr-Nb à 2,5% dans le domaine 10^{-7} h^{-1} mais réduisait sensiblement la vitesse de fluage dans les domaines de 10^{-5} h^{-1} et au-dessus. L'influence de l'effort sur la vitesse de fluage en réacteur semble obéir à une loi en puissance dans le Zircaloy-2 écroui à froid et l'alliage Zr 2,5% Nb avec un exposant de l'effort égal à 3 environ. L'influence de la température sur la vitesse de fluage en réacteur est modifiée autour de 350 °C à la fois dans le Zircaloy-2 écroui à froid et l'alliage Zr-2,5% Nb traité thermiquement. Au-dessus de 350 °C, l'influence de la température peut être exprimée par une énergie d'activation apparente de 50–60 000 cal/mole, tandis qu'en dessous de 350 °C, la valeur de l'énergie d'activation chute à 20 000 cal/mole environ.

Der Bericht fasst die experimentellen Ergebnisse von 24 gleichachsigen Kriechversuchen bei Bestrahlung von verschiedenen Zirkonlegierungen zusammen. Die Spannungen liegen dabei im Bereich von 7 bis 42 kg/mm² und die Temperaturen zwischen 200 und 400 °C. Für kaltbearbeitete Zirkaloy-2-Proben bei schnellen Neutronen (Flüsse von $0.4–1.2 \times 10^{13}$ n/cm² sec, $E > 1$ MeV) steigt die Kriechgeschwindigkeit bei 300 °C um bis zu einer Grössenordnung an. Die durch die Bestrahlung hervorgerufenen Anstiege waren kleiner bei kaltbearbeitetem und warmbehandeltem Zr-2.5 Gew. % Nb und nicht messbar in geglühtem Zirkon. Vorherige Bestrahlung hatte keinen bedeutenden Einfluss auf das Kriechen von Zirkaloy-2 und Zr-2.5 Gew. % Nb bei etwa 10^{-7} h^{-1}, verminderte aber die Kriechgeschwindigkeit bei 10^{-5} h^{-1} und in höheren Bereichen wesentlich. Die Spannungsabhängigkeit der Kriechgeschwindigkeit bei Bestrahlung scheint einem Exponentialgesetz bei kaltbearbeitetem Zirkaloy-2 und Zr-2.5 Gew. % Nb zu folgen mit einem Spannungsexponenten von ungefähr 3. Die Temperaturabhängigkeit wechselt um 350 °C sowohl für kaltbearbeitetes Zirkaloy-2 als auch für warmbehandeltes Zr-2.5 Gew. % Nb. Oberhalb von 350 °C kann die Abhängigkeit durch eine Aktivierungsenergie von 50–60 000 cal/mol ausgedrückt werden, während unterhalb von 350 °C der Wert auf etwa 20 000 cal/mol abfällt.

Table 1

The heat treatment, hardness and tensile properties of zirconium alloys tested for creep in-reactor.

Material (code no.)	Material	Heat treatment	Hardness on transv. section (VHN)	Tensile properties (longitudinal sections)								In-reactor creep test no.	Average grain size (μm)
				Room temperature				300 °C					
				0.2% Y.S. (kg/mm²)	UTS (kg/mm²)	Total† eln. (%)	R.A. (%)	0.2% Y.S. (kg/mm²)	UTS (kg/mm²)	Total† eln. (%)	R.A. (%)		
9	Zircaloy-2 rod	20% cold-worked, stress-relieved 72 h at 400 °C	230	49.9	61.4	12	30	35.9	37.3	7	58	R-2	7
11	Zircaloy-2 tube	19% cold-worked, autoclaved 72 h at 400 °C	191	47.1	61.2	19		33.0	39.4	20		R-8, R-9	7
18	Zircaloy-2 rod	20% cold-worked, stress-relieved 72 h at 400 °C	227	62.6	69.6	18	39	32.3	35.2	20	47	R-4, Rx-20	8
25	Zircaloy-2 tube	16% cold-worked, autoclaved 72 h at 400 °C	193	49.9	60.5	13	34	31.6	35.9	12	45	R-6, Rx-14, Rx-21	7
27	Zr-2.5 wt % Nb rod	water quenched from 850 °C aged 24 h at 500 °C	268	76.6	84.4	14	60	57.7	65.4	15	80	R-7, Ru-6, Ru-7	
33	Zirconium rod	annealed	66	12.7	19.7	50	72	8.4	14.1	36	92	Rx-19	98

34	Zr-2.5 wt % Nb tube	21% cold-worked, autoclaved 72 h at 400 °C	214	54.8	78.0	14	44	37.3	52.0	14	56	R-11, R-12 Rx-17
36	Zr-2.5 wt % Nb rod	water quenched from 875 °C, cold-drawn 30% aged 24 h at 500 °C	247	60.5	82.3	19	53	47.1	53.4	19	68	Rx-13, Ru-1
37	Zr-2.5 wt % Nb tube	water quenched from 880 °C cold-drawn 22% aged 24 h at 500 °C	252	60.5	74.5	23	54	40.8	52.7	20	61	Rx-16
39	Zr-2.5 wt % Nb tube	water quenched from 880 °C, cold-drawn 5%, aged 24 h at 500 °C	255	67.5	80.9	19	52	42.2	57.7	19	62	Rx-18
42	Zircaloy-2 tube	19% cold-worked, autoclaved 72 h at 400 °C	205	57.0	61.4	14	36	32.3	38.7	17		Rx-15
50	Zr-2.5 wt % Nb tube	water quenched from 870 °C cold-drawn 16%, aged 24 h at 500 °C	262	68.2	85.8	20	51	52.0	61.9	17	65	Rx-22
51	Zr-2.5 wt % Nb tube	water quenched from 870 °C, cold-drawn 5%, aged 24 h at 500 °C	261	68.9	84.4	18	55	50.6	59.1	17	53	Rx-24
52	Zr-2.5 wt % Nb tube	water quenched from 870 °C, cold-drawn 11% aged 24 h at 500 °C	274	68.2	85.1	18	47	54.1	61.9	18	51	Rx-23, Ru-5

† 2.5 cm gauge length.

1. Introduction

Zirconium alloys are widely used in the nuclear industry. Because of the excellent corrosion resistance and low neutron absorption cross-section Zircaloy-2 has been chosen as the main structural material for the reactor core of Canadian CANDU-type power reactors. The creep rate of Zircaloy-2 is, however, increased by a fast neutron flux in the temperature range 220–375 °C. Investigations have been in progress at Chalk River since 1960 to determine whether or not a stronger zirconium alloy is available which is less affected by neutron flux than Zircaloy-2, and to study the effect of flux on creep. Of the alloys investigated the heat treatable zirconium-niobium alloys seem to be

TABLE 2

Ingot analyses of materials tested. Alloy composition (wt %).

Element	Material (code no., table 1)										
	9	11	18	25	27	33	34	36	37, 39	42	50-51-52
Sn	1.36	1.45	1.57	1.43	–	–	–	–	–	1.56	–
Fe	0.13	0.13	0.13	0.13	–	–	–	–	–	0.13	–
Cr	0.10	0.11	0.10	0.11	–	–	–	–	–	0.11	–
Ni	0.05	0.05	0.06	0.06	–	–	–	–	–	0.06	–
Nb	–	–	–	–	2.70	–	2.75	2.51	2.53	–	2.47
Zr	Balance										
	Impurity content (ppm)										
Al	38	65	<30	20	46	< 25	49	44	46	–	49
B	< 0.3	< 0.2	< 0.5	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	–	< 0.2
C	90	187	242	340	65	35	120	148	90	–	178
Cd	< 0.2	< 0.5	< 0.5	< 0.5	< 0.3	< 0.3	< 0.3	< 0.3	< 0.2	–	< 0.3
Co	< 10	< 5	< 5	<20	< 5	< 5	< 5	< 5	< 5	–	< 5
Cr	–	–	–	–	25	36	123	115	104	–	< 10
Cu	< 25	25	24	36	30	< 25	<25	25	25	20	< 25
Fe	–	–	–	–	960	135	1219	860	890	–	1330
H	45	22	23	–	5	3	11	7	8	13	8
Hf	< 100	71	<45	95	67	113	<40	45	74	–	56
Mg	< 10	<10	<15	<20	< 10	< 10	<10	< 10	< 10	–	< 10
Mn	35	20	<10	<20	< 10	< 10	11	15	< 10	–	< 10
Mo	< 20	<10	<20	<20	< 10	< 10	<10	12	< 10	–	< 10
N	35	40	45	48	24	23	26	59	39	42	56
Nb	–	–	–	–	–	<100	–	–	–	–	–
Ni	–	–	–	–	40	10	25	13	< 10	–	< 10
O	<1200	600	1540	–	1175	70	1200	1300	935	1250	890
Pb	< 20	10	<15	<20	< 5	7	< 5	10	< 5	–	< 5
Si	77	70	31	35	94	< 40	50	58	73	–	56
Sn	–	–	–	–	< 10	< 10	<10	< 10	< 10	–	< 10
Ta	–	–	–	–	<200	<200	–	<200	<200	–	<200
Ti	< 25	<30	<20	<20	20	< 20	<50	< 50	< 50	–	< 50
U	–	–	–	1	< 0.5	< 0.5	< 0.5	< 0.5	–	–	< 0.5
V	< 25	<20	<20	<20	< 5	5	5	< 5	< 5	–	< 5
W	< 21	60	<20	<20	< 25	< 25	<25	50	42	–	< 25
Zn	< –	–	<10	–	< 50	< 50	–	< 50	< 50	–	< 50

the most promising. This report summarizes the uniaxial creep experiments which have been completed.

2. Material

Of the 14 different batches of material investigated one consisted of crystal bar zirconium, five Zircaloy-2 and eight Zr-2.5 wt % Nb. The heat treatment and mechanical properties of each batch of material are given in table 1 and their chemical ingot analyses in table 2. The creep specimens were machined either from 1.3 cm dia. bar stock (materials 9, 18, 27, 33 and 36) or longitudinal pressure tube sections. Because of the pressure tube wall thickness (0.32 to 0.40 cm), the specimen gauge diameter could not be made greater than 0.25 cm and for most specimens was chosen as 0.20 cm. The average number of grains in a cross-section of Zircaloy-2 specimens was about 250, and in annealed zirconium about 20.

The textures of both the rods and the pressure tubes have been reported by Cheadle and Ells [1,2]. The zirconium grains were oriented in the rods and similarly in the tubes so that the prismatic $\{10\bar{1}0\}$ plane normals were predominantly parallel to the stress axis of the creep specimens.

The metallographic structures of the specimens before and after creep are described in detail elsewhere [3]. Pertinent structural information obtained [4-6] from transmission electron microscopy is summarized in table 3.

3. Experimental procedure

The creep tests were carried out inside hollow fuel rods in the NRX and NRU reactors. The fast neutron flux ranged between 5 and 12×10^{12} n/cm^2·sec ($E>1$ MeV) in NRX and between 2 and 3×10^{13} n/cm^2·sec in NRU.

The in-reactor creep machines have been described in detail elsewhere [7,8]. The stress on the creep specimens was applied by means of a pressurized bellows and maintained constant to $\pm$ 0.2%. The strain was measured with a pneumatic gauge of 25×10^{-6} cm sensitivity. The specimen gauge length was 5 cm and the dia. 0.20 cm. A test rig of 1×10^{-4} cm sensitivity

TABLE 3

Summary of microstructure of zirconium alloys tested, as obtained from electron microscope examinations.

Material	Principal characteristics of structure prior to testing	Reference
Cold worked Zircaloy-2	Deformed equiaxed α grains, containing dislocation density $\sim10^{10}$ lines/cm^2. Crude cellular structure of diameter ~1 μm. After irradiation with fast neutrons to an integrated flux of 10^{20} n/cm^2, dislocation loops were observed, but over-all dislocation form and distribution was little changed.	[3]
Cold worked Zr-2.5 wt % Nb	Grain size smaller than for Zircaloy-2, but dislocation configuration similar.	
Quenched and aged Zr-2.5 wt % Nb	Equiaxed α in matrix of martensitic α'. Small Nb-rich precipitates distributed throughout the martensitic phase. Dislocation density low, $\sim10^7$ lines/cm^2.	[4]
Quenched/cold worked/aged Zr-2.5 wt % Nb	Effect of intermediate cold work>10% reduction is to cause recrystallization in the α' phase during aging. Retained α' after aging is $\sim60\%$ with 20% cold reduction and $\sim10\%$ with 30% cold reduction. Thus with increasing cold work the aged structure contains progressively more α. Dislocation densities after aging at 500 °C for 24 h increase from 10^7 lines/cm^2 without intermediate coldwork to $\sim10^9$ lines/cm^2 after 10–20% coldwork and fall to 10^7–10^8 lines/cm^2 after 30% coldwork as recrystallization proceeds.	[5]

TABLE 4

In-reactor creep tests on Zircaloy-2.

Test no.	Material (code no., table 1)	Test conditions: Temperature (°C)	Test conditions: Stress (kg/mm²) (lb/in²)	Test conditions: Fast flux ($E>1$ MeV) (n/cm²·sec) $\times 10^{12}$	Loading strain (%)	Total test time (h)	Properties at given test conditions: Duration (h)	Properties at given test conditions: Total strain near end of test (%)	Properties at given test conditions: Creep rate near end of test (h^{-1})	Creep rate of laboratory control test after same total time as in-reactor test (h^{-1})	Remarks (in-reactor tests only)
R-2	9	300	21.1 (30 000)	5.0	0.185	2000	2000	0.605	$1.8\times10^{-6}\pm30\%$	$0.2\times10^{-6}\pm30\%$	Test in air. Temperature dropped frequently to 100 °C
R-2	9	300	21.1	6.6	0.185	2300	300	0.765	$1.5\times10^{-6}\pm10\%$	$<0.2\times10^{-6}$	Machine moved to a higher flux position
R-4	18	300	14.1 (20 000)	5.9	0.220	260	260	0.275	$6\times10^{-7}\pm30\%$	$6\times10^{-7}\pm20\%$	All test 4 in air
R-4	18	300	14.1	0	0.250	880	620	0.305	$0.8\times10^{-7}\pm30\%$	$0.9\times10^{-7}\pm20\%$	Test moved to storage facility out of reactor
R-4	18	300	14.1	5.8	0.310	1280	400	0.350	$3.5\times10^{-7}\pm30\%$	$0.4\times10^{-7}\pm40\%$	Test back in reactor. At temperature for 140 h before reloading
R-4	18	300	21.1 (30 000)	0	0.370	1900	620	0.450	$4.0\times10^{-7}\pm20\%$	$4.0\times10^{-7}\pm20\%$	Test in storage facility out of reactor
R-4	18	300	21.1	3.6	0.460	3230	1330	0.620	$8.0\times10^{-7}\pm10\%$	$2.0\times10^{-7}\pm30\%$	Test moved back into reactor. Creep rate approximately constant
R-4	18	300	21.1	0	0.585	5950	2720	0.640	$0.4\times10^{-7}\pm100\%$	$0.3\times10^{-7}\pm100\%$	Test in storage facility. Temperature deliberately cycled down by 50 °C during last 1200 h. No measurable effect
R-4	18	300	21.1	4.2	0.655	6920	970	0.715	$7.0\times10^{-7}\pm20\%$	-	Reactor shutdown approximately one third of the duration
R-6	25	300	14.1 (20 000)	5.4	0.210	3600	3600	0.360	$3.5\times10^{-7}\pm10\%$	$0.4\times10^{-7}\pm20\%$	Creep rate steady
R-6	25	300	14.1	0	0.210	3850	250	0.363	$0.5\times10^{-7}\pm10\%$	$0.4\times10^{-7}\pm20\%$	During prolonged reactor shutdown
R-6	25	300	14.1	6.0	0.210	5735	1885	0.405	$2\text{–}3.5\times10^{-7}$	$0.2\times10^{-7}\pm50\%$	Non-steady operation of reactor. Many shutdowns
R-6	25	320	14.1	6.0	0.400	5783	48	0.405	-	-	Unable to determine creep rate because of scatter
R-6	25	350	14.1	6.0	0.400	5882	99	0.420	$1.3\times10^{-6}\pm100\%$	$1.1\times10^{-6}\pm10\%$	
R-6	25	375	14.1	6.0	0.410	5930	48	0.455	$1.0\times10^{-5}\pm10\%$	$6.4\times10^{-5}\pm20\%$	
R-6	25	390	14.1	6.0	0.450	5985	55	0.56	$2.0\times10^{-5}\pm20\%$	$1.6\times10^{-5}\pm10\%$	
R-8	11	300	11.6 (16 500)	0	0.148	35 040	35 040	0.240	$5\times10^{-9}\pm50\%$	-	Test out of reactor
R-8	11	300	11.0 (15 700)	6.0	0.228	35 440	400	0.255	$4\times10^{-7}\pm50\%$	-	Test moved to reactor. Had to be terminated because of loss of stress. Creep rate still decreasing

R-9	11	300	12.7 (18 000)	0	0.173	10 000	10 000	0.241	$3\times10^{-8}\pm75\%$	-	Test out of reactor
R-9	11	300	7.7 (11 000)	6.1	0.105	12 140	2140	0.117	$5\times10^{-8}\pm60\%$	-	Test moved to reactor
R-9	11	300	12.7 (18 000)	5.8	0.179	13 620	1480	0.245	$3\times10^{-7}\pm20\%$	-	Creep rate steady
Rx-14	25	300	14.1 (20 000)	7.3	0.205	850	850	0.280	$5\times10^{-7}\pm20\%$	$1.4\times10^{-7}\pm10\%$	Specimen pre-irradiated to 3×10^{20} n/cm² at 300 °C before start of test
Rx-14	25	300	14.1	6.4	0.205	2000	1150	0.310	$3\times10^{-7}\pm30\%$	$0.8\times10^{-7}\pm20\%$	Creep rate steady
Rx-14	25	320	14.1	6.8	0.310	2270	270	0.327	$6\times10^{-7}\pm20\%$	-	
Rx-14	25	350	14.1	6.8	0.335	2480	210	0.360	$1.4\times10^{-6}\pm20\%$	(see test R-6 control)	Stress off for 60 h, early in the period
Rx-14	25	375	14.1	6.8	0.367	2513	33	0.403	$1.0\times10^{-5}\pm10\%$	(see test R-6 control)	
Rx-14	25	400	14.1	6.8	0.408	2543	30	0.508	$2.7\times10^{-5}\pm10\%$		
Rx-14	25	400	14.1	0	0.508	2576	33	0.553	$1.3\times10^{-5}\pm10\%$		During shutdown, creep rate decreasing
Rx-15	42	220	21.1 (30 000)	9.6	0.272	2130	2130	0.365	$1.5\times10^{-7}\pm30\%$	$<1\times10^{-8}$	
Rx-15	42	260	21.1	9.3	0.384	3010	880	0.420	$5.0\times10^{-7}\pm40\%$	$4\times10^{-8}\pm30\%$	Results uncertain because of strain gauge instability
Rx-15	42	300	21.1	9.4	0.420	4300	1290	0.500	$8\times10^{-7}\pm30\%$	$7\times10^{-8}\pm30\%$	Results uncertain because of strain gauge instability. Specimen temperature low for considerable period of time
Rx-15	42	350	21.1	9.4	0.500	4826	526	0.66	$3.2\times10^{-6}\pm10\%$	$1.5\times10^{-6}\pm10\%$	Creep rate steady
Rx-15	42	350	21.1	0	0.500	4450	120	0.530	$1.4\times10^{-6}\pm20\%$	-	During reactor shutdown
Rx-15	42	375	21.1	0	0.70	4880	54	0.76	$1.3\times10^{-5}\pm10\%$	$8\times10^{-6}\pm20\%$	During reactor shutdown
Rx-15	42	300	21.1	9.4	0.76	4948	68	0.77	-	-	Temperature lowered to preserve strain gauge
Rx-15	42	375	21.1	9.4	0.82	4977	29	0.87	$2\times10^{-5}\pm10\%$	$8\times10^{-6}\pm20\%$	Creep rate steady
Rx-15	42	400	21.1	9.4	0.90	4985	8	0.95	$7\times10^{-5}\pm10\%$	$7\times10^{-5}\pm20\%$	Creep rate steady
Rx-20	18	350	31.6 (45 000)	11.6	0.58	4.4	4.4	2.37	$5.0\times10^{-3}\pm10\%$	Failed on loading	After 4.4 h stress lowered to 1.8 kg/mm² (2 500 lb/in²). Specimen kept at 350 °C for 100 h before reloading to 21.1 kg/mm² (30 000 lb/in²)
Rx-20	18	350	21.1 (30 000)	11.6	2.13	382	280	2.58	$6.0\times10^{-6}\pm10\%$		Creep rate steady
Rx-21	25	300	31.6 (45 000)	0.6	0.43	305	305	0.87	$1.0\times10^{-5}\pm20\%$	9×10^{-5} (ruptured after approximately 200 h -mean of three tests)	Specimen pre-irradiated to 3×10^{20} n/cm² at 300 °C. Test interrupted after 305 h and moved to a higher flux position
Rx-21	25	300	31.6	8.4	0.86	1085	780	1.56	$1.2\times10^{-5}\pm10\%$		Creep rate steady. Heater failed after 1085 h
Rx-21	25	300	31.6	0	0.86	660	100	1.18	$0.3\times10^{-5}\pm10\%$		During reactor shutdown
Rx-21	25	170–180	31.6	8.4	0.86	1250	160	1.55	$0.14\times10^{-5}\pm30\%$		Temperature fluctuated with variation in nuclear heatings. Test removed after 1250 h for heater repairs, reinstalled 50 h later
Rx-21	25	300	31.6	8.4	1.52	1590	290	1.93	$1.2\times10^{-5}\pm10\%$		Creep rate steady
Rx-21	25	300	28.1 (40 000)	8.4	1.92	1655	65	1.95	$3.5\times10^{-6}\pm50\%$		Specimen unloaded after reaching the end of strain range (1.95%)

Table 5

In-reactor creep of Zr

Test no.	Material (code no.)	Test conditions			Loading strain (%)	Total test time (h)
		Temperature (°C)	Stress kg/mm² (lb/in²)	Fast flux $E > 1$ MeV (n/cm²·sec) $\times 10^{12}$		
R-7	27	300	21.1 (30 000)	6.2	0.240	835
R-7	27	300	21.1	6.2	0.275	2650
R-11	34	300	16.2 (23 000)	7.0	0.254	3200
R-12	34	300	11.6 (16 500)	5.6	0.102	2800
Rx-13	36	300	16.2 (23 000)	5.7	0.226	2569
Rx-13	36	350	16.2	5.7	0.424	2612
Rx-13	36	400	16.2	5.7	0.450	2629
Rx-16	37	300	16.2	5.7	0.288	295
Rx-16	37	300	25.6 (36 500)	5.7	0.375	1750
Rx-16	37	300	31.6 (45 000)	5.7	0.558	2980
Rx-16	37	320	31.6	5.7	0.628	3130
Rx-16	37	300	11.2 (16 000)	5.7	0.388	5540
Rx-17	34	300	14.1 (20 000)	6.8	0.249	2125
Rx-18	39	300	11.2 (16 000)	5.3	0.232	2100
Rx-22	50	300	11.2	5.8	0.184	4900
Rx-23	52	260	14.1 (20 000)	5.3	0.195	5300
Rx-23	52	300	14.1	5.3	0.27	6250
Rx-23	52	350	14.1	5.3	0.31	6620
Rx-24	51	300	42.2 (60 000)	8.9	0.568	930
Ru-1	36	300	16.2 (23 000)	24	0.218	2560
Ru-5	52	300	11.2 (16 000)	22	0.266	1120
Ru-6	27	300	11.2	24	0.182	1400
Ru-7	27	300	11.2	21	0.210	3140

TABLE 5

-2.5 wt % Nb alloys.

Properties at given test conditions			Creep rate of laboratory control test after same total time as in-reactor test (h^{-1})	Remarks (In-reactor tests only)
Duration (h)	Total strain near end of test (%)	Creep rate near end of test (h^{-1})		
835	0.293	$1.0\times10^{-7}\pm30\%$	1.7×10^{-7}	Heater failed. Specimen retained in reactor at about 150 °C, unstressed for 600 h before reloading
1210	0.301	$0.8\times10^{-7}\pm30\%$	2.0×10^{-7}	Test terminated because of repeated heater failure
3200	0.355	$2.0\times10^{-7}\pm15\%$	4.0×10^{-8}	Specimen temperature excursion to 400 °C after 2800 h resulted in a strain step of 0.02%
2800	0.157	$7.0\times10^{-8}\pm10\%$	5.0×10^{-8}	Creep rate steady
2569	0.420	$2.0\times10^{-7}\pm15\%$	3.5×10^{-7}	Specimen temperature excursion to 350 °C after 750 h resulted in a strain step of 0.08%
43	0.439	$1.5\times10^{-6}\pm10\%$	7.0×10^{-6}	
17	0.520	$4.3\times10^{-5}\pm10\%$	1.3×10^{-4}	
295	–	–	–	Strain gauge unstable, raised stress to overcome instability
1455	0.478	$1.5\times10^{-7}\pm30\%$	6.0×10^{-7}	
1230	0.628	$2.0\times10^{-7}\pm30\%$	6.0×10^{-7}	Stress lowered for about 50 h around 1900 and 2650 h
150	0.635	–	–	Specimen temperature raised to overcome strain gauge instability.
2410	0.358	$9.5\times10^{-8}\pm30\%$	7.0×10^{-8}	Specimen recovered creep strain for the first 1000 h down to 0.346%
2125	0.340	$1.05\times10^{-7}\pm40\%$	1.5×10^{-7}	
2100	0.295	$0.80\times10^{-7}\pm40\%$	0.9×10^{-7}	
4900	0.308	$9.0\times10^{-8}\pm20\%$	$<1\times10^{-8}$	
5300	0.27	$1.05\times10^{-7}\pm15\%$	2×10^{-8}	Creep rate steady
950	0.30	$4\times10^{-7}\pm20\%$	7×10^{-8}	Creep rate steady
370	0.35	$1.3\times10^{-6}\pm20\%$	1.4×10^{-6}	Creep rate steady
930	0.755	$8.0\times10^{-7}\pm15\%$	2×10^{-6}	
2560	0.648	$1.4\times10^{-6}\pm20\%$	1.3×10^{-7}	Frequent temperature cycling downward and occasionally up to 400 °C. Stress off for 100 h after 2000 h
1120	0.420	$1.15\times10^{-6}\pm15\%$	7.0×10^{-8}	
1400	1.260	$8.0\times10^{-7}\pm50\%$	0.5×10^{-7}	Results uncertain because of scatter. Temperature excursion to 450 °C after 430 h caused a strain step of 0.76%
3140	0.440	$1.8\times10^{-7}\pm50\%$	$<0.5\times10^{-7}$	Results uncertain because of scatter. Temperature cycled down after 600 and 1500 h

TABLE 6

In-reactor creep tests of annealed zirconium.

Temp. (°C)	Test conditions: Stress kg/mm² (kpsi)	Test conditions: Fast flux ($E>1$ MeV) (n/cm²·sec) $\times 10^{12}$	Loading strain (%)	Total strain (%)	Duration at any given condition (h)	Accumulated test time (h)	Creep rate near end of test (h^{-1})	Creep rate of laboratory test after same total time as in-reactor test (h^{-1})	Remarks
200	7.0 (10)	6.7	0.17	0.23	2100	2100	$5–10\times10^{-8}$	1.5×10^{-7}	Large scatter of test data beyond 1700 h. Machine removed from reactor for repairs and returned for test at higher stress and temperature on the same specimen
220	14.1 (20)	6.1	0.35	0.78	1500	3600	4.0×10^{-7}	3×10^{-7}	Suffered large temperature cycles in the early part of 220 °C test
220	14.1	0 (during shutdown)		0.77	100	3400	$4–5\times10^{-7}$		
260	14.1	6.1	0.80	0.98	300	3950	4×10^{-6}	6×10^{-5}	Creep rate steady
260	10.5 (15)	6.1	0.92	0.96	300	4250	1.6×10^{-6}	1.4×10^{-6}	Creep rate steady
260	10.5	(during shutdown)		0.93	150	4100	1.6×10^{-6}		
300	10.5	,,	0.97	1.02	35	4275	2.2×10^{-5}		
300	10.5	6.1		1.14	25	4300	2.2×10^{-5}	–	
300	7.0	6.1	1.105	1.12	60	4360	2.7×10^{-6}	–	

was used to measure the specimen gauge length before and after a creep test thereby providing a check on the results obtained with the pneumatic gauge. The specimen temperature was controlled by a multisection resistance heater. Except for tests R-2, R-4 and Ru-1 which were subject to frequent downward temperature cycles because of inadequate temperature control, the specimen temperature was maintained constant to ± 2 °C during normal reactor operation and even during reactor outages the temperature dropped only by about 5 °C for a duration of approximately one minute. The temperature difference along the specimen gauge length did not exceed 3 °C. The tests R-2 and R-4 were carried out in air, the remainder in an atmosphere of helium.

During three tests (R-2, R-4 and Rx-21) the creep machines were moved to different positions of the reactor, or to a storage facility outside the reactor, to determine the effect of different neutron fluxes on the creep rate; also, in the R-4 test, to compare material under irradiation with the same material in an "as-irradiated" state.

At low creep rates in-reactor specimens required 2000–4000 h to reach steady values. To save time a number of specimens were tested at more than one stress and temperature once steady state conditions had been reached.

The average scatter of experimental points ranged between 0.002 and 0.010% strain. This was primarily caused by variations in reactor power and associated differential expansions of

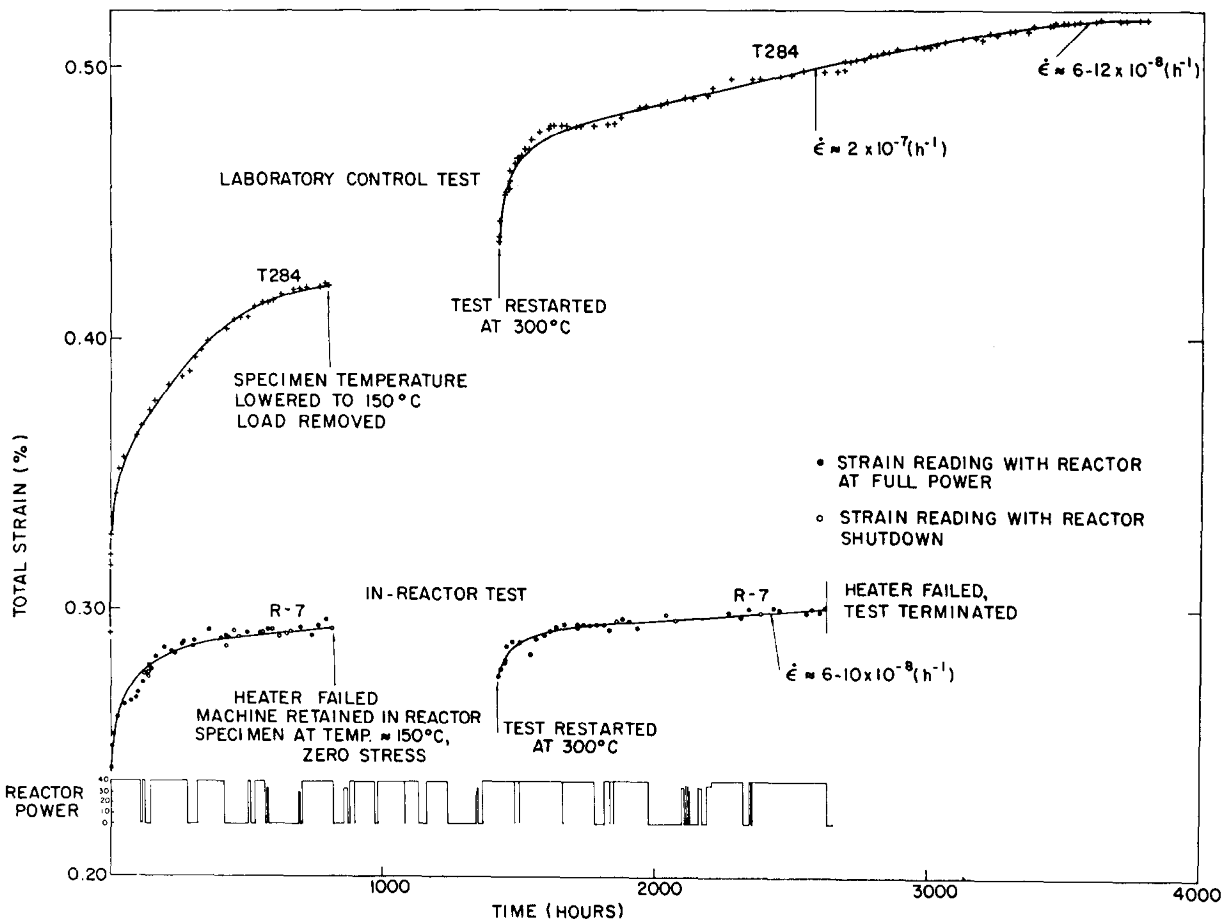

Fig. 1. In-reactor creep of Zr-2.5 wt % Nb (water quenched from 850 °C, aged 24 h at 500 °C). Stress: 21.1 kg/mm² (30 000 psi); Temp.: 300 °C; neutron flux: 6×10^{12} n/cm²·sec ($E >$1MeV).

creep machine components. On some occasions scatter resulted from instabilities in the pneumatic strain gauge over a narrow range of strain. Because of scatter, 1000 h or more were sometimes required to define low creep rates (below 10^{-7}/h) to an accuracy of $\pm$ 50%. Unless the measured creep rates were 10^{-6}/h or higher, even a factor of 10 change in the creep rate due to neutron flux could not be verified during the normal reactor shutdown periods of 100–200 h duration.

The unirradiated control tests were conducted in both standard dead weight creep machines manufactured by Instron Corporation and laboratory models of the in-reactor creep machine. The strain measurement on the Instron machines was made with mechanical extensometers. No significant difference in the creep rate was observed between tests carried out in the two different types of machines, however, there was some difficulty in obtaining reproducible loading strains inside the Instron creep machines.

4. Results

The test results are summarized in tables 4, 5 and 6. Examples of in-reactor creep curves are shown later in figs. 1–3, 5, 7. The details on each of the tests are described elsewhere [3]).

Neutron irradiation increased the creep rate of cold-worked Zircaloy-2 in the temperature range 200–350 °C by an order of magnitude or more; the creep rate of cold-worked Zr-2.5 wt % Nb at 300 °C was increased to a lesser extent. The in-reactor test R-7 on quenched and aged Zr-2.5 wt % Nb at 300 °C had a lower loading strain and creep rate than its laboratory control tests. The low value of the loading strain in test R-7 is considered to result from differences in the elastic moduli of the two specimens and radiation induced hardening which reduced the plastic deformation during stressing (the R-7 specimen was preirradiated for one week prior to stressing). The lower value of the in-reactor creep rate in fig. 1 is not considered to represent the final, steady state condition in this material. A reversal in the relative magnitude of creep rate can occur in time because of continuously decreasing creep rate out-of-flux, while the in-reactor rate stays constant. From comparison with the long time laboratory data (fig. 1) the effect of flux in test R-7 is small.

In Zr-2.5 wt % Nb with cold-work between quenching and aging treatments the effect of flux on creep varied with the applied stress and to some extent with the degree of cold-work. The test specimens with 30% intermediate cold-work showed an effect of flux (fig. 2), similar to that in cold-worked Zircaloy-2. The results of test Ru-1 which show a very large effect of flux on creep are suspect because of frequent temperature cycling. The laboratory control test with the same type of temperature cycling has not yet been carried out. As noted in table 3 electron microscopic investigations revealed that extensive recrystallization had occurred during the 500 °C aging treatment, the twinned martensitic α' phase that characterises the quenched and aged material being replaced by small subgrains of α. The tests with material subjected to less intermediate cold-work showed only a small effect of flux at stresses above 14.1 kg/mm^2. At stresses below 14.1 kg/mm^2 the creep rates were difficult to determine accurately because of the scatter due to differential expansions in the machine. For the two tests that lasted 5000 h (Rx-22 and Rx-23) the creep rates could be determined to $\pm$ 20% and appeared to be steady, while for the shorter ones (Rx-16, Rx-18) the uncertainty amounted to $\pm$ 50%. The results suggested an irradiation effect on creep similar in magnitude to that in cold-worked Zircaloy-2. Furthermore the creep rates appeared to be independent of the degree of intermediate cold-work in the range 5–22%.

Three creep tests (Rx-14, 20 and 21) were carried out on specimens that were irradiated to an integrated dose of 3×10^{20} n/cm^2 ($E>1$ MeV) at 300 °C prior to creep testing. The effect on creep rate varied with the material and test conditions. At low stresses the effect appeared to be negligible, but at high stresses

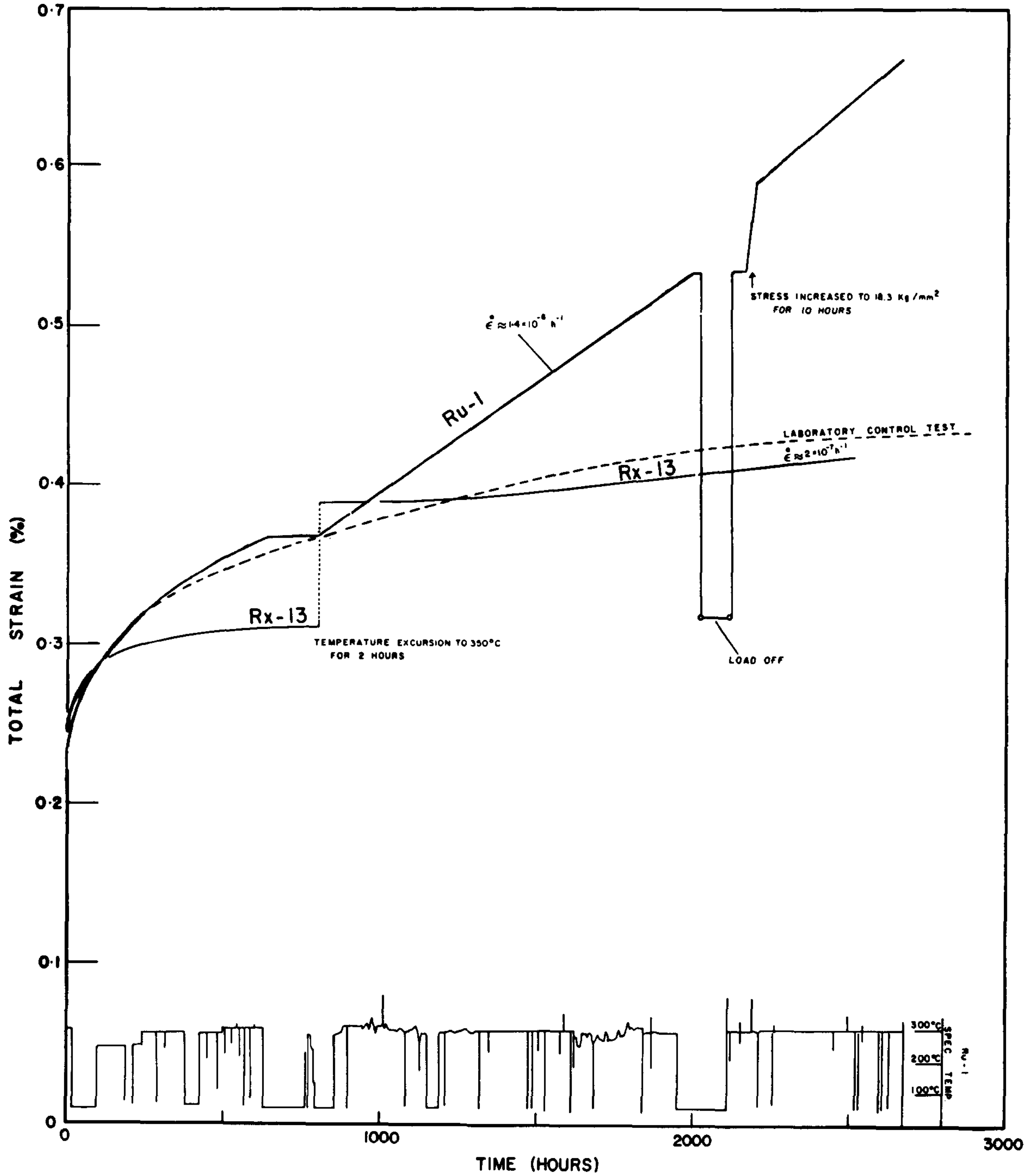

Fig. 2. In-reactor creep of Zr-2½ wt % Nb at 300 °C and 16.2 kg/mm² (23 000 psi). Material: 880 °C quenched, 30% cold-drawn, 24 h at 500 °C tempered rod specimen. Test Ru-1 in NRU reactor, fast flux approximately 2.4×10^{13} n/cm²·sec ($E > 1$ MeV). Test: Rx-13 in NRX reactor, fast flux approximately 0.57×10^{13} n/cm²·sec ($E > 1$ MeV).

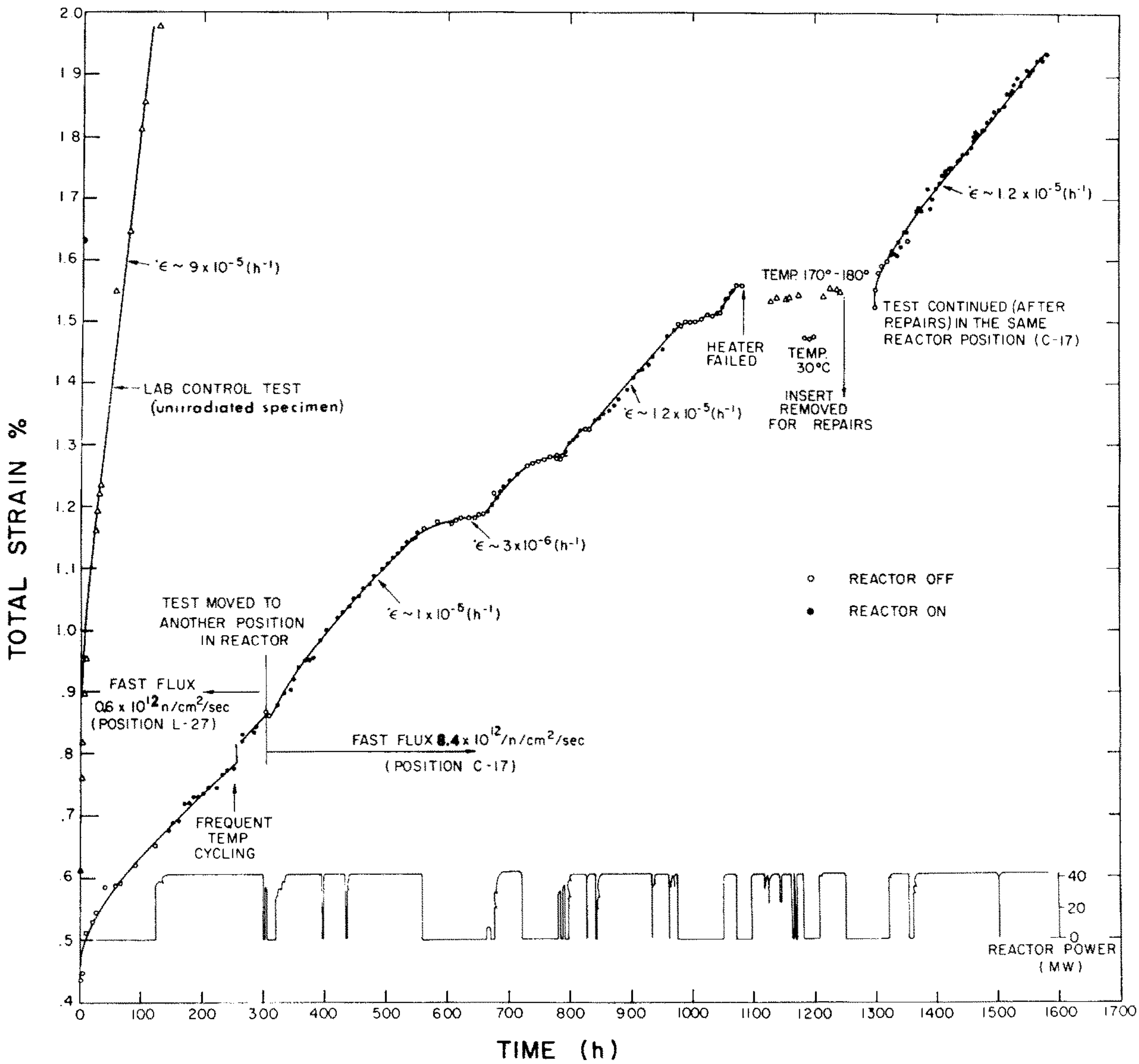

Fig. 3. In-reactor creep test Rx-21 (in NRX) on 16% cold-worked, autoclaved Zircaloy-2 pressure tube specimen (pre-irradiated to 3×10^{20} n/cm² ($E>1$ MeV) at 300 °C). Test conditions: 31.6 kg/mm² (45 000 psi) – 300 °C; neutron flux: 0.6 to 8.4×10^{12} n/cm²·sec ($E>1$ MeV).

the preirradiated specimens had considerably lower creep rates than the laboratory control tests (fig. 3).

5. Discussions of results

5.1. General

There is very little published information available on the effect of neutron irradiation on creep of zirconium alloys or metals in general. When present the irradiation effect manifested itself largely by a steady creep rate after an initial period of time that varied with material, stress and flux intensity. This behaviour is in contrast with the transient, continuously decreasing creep rate obtained in the laboratory tests (fig. 4). Thus the difference between the in-reactor and laboratory creep rates changed with time and even reversed itself in those tests for which the laboratory rate happened to be higher initially.

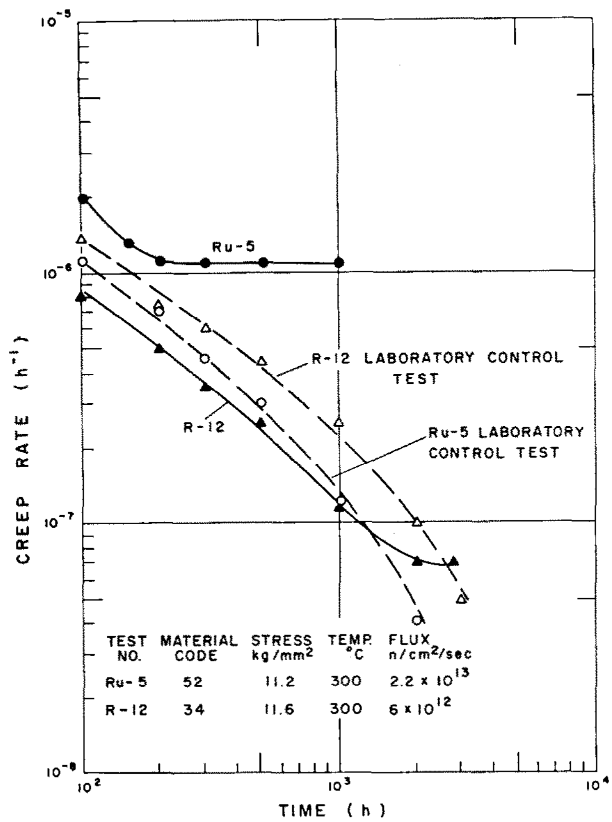

Fig. 4. Variation of in-reactor and laboratory creep rates with time of two Zr-2.5 wt % Nb alloys.

Comparisons are made in the text between laboratory control tests that were carried out on unirradiated materials and corresponding in-reactor tests. Because of structural changes introduced by the irradiation, the comparisons may not be valid. From the similar creep curves of Zircaloy-2 at 14.1 kg/mm² and 300 °C in the reactor on both unirradiated and pre-irradiated specimens and the laboratory test on unirradiated specimens, the presence of irradiation induced defects does not seem to have a large effect on the creep rate at low stresses (tests R-6, Rx-14 in table 4). At higher stresses the in-reactor creep rates of preirradiated materials are an order of magnitude lower than in the laboratory (on unirradiated material) because the radiation induced obstacles are more effective in blocking the movement of dislocations [9]). This type of behaviour is demonstrated in tests Rx-19, Rx-20 and Rx-21 (table 4, figs. 3 and 5), where the preirradiated specimens easily supported the applied high stress while the unirradiated laboratory control tests failed on loading or soon thereafter.

The irradiation induced hardening may in part be responsible for the different irradiation effect on creep of annealed zirconium reported by Faris [10]). He quoted the creep data of Fillnow, Cook and Johnson on zirconium as having up to 20 times lower creep rate at 260 °C under irradiation than either before or after irradiation, while at Chalk River no measurable effect was detected in the temperature range 200 to 300 °C. Faris did not give any experimental details to allow an assessment of the reliability of the data by Fillnow et al. The tests seem to have been of very short duration (3 to 30 h) and probably had not reached steady state conditions. In the Chalk River test at 7 kg/mm² stress and 200 °C there was no significant difference between the in-reactor and laboratory tests (fig. 5), nor was there any marked change in creep rate during reactor shutdowns. However, when the stress was raised from 7.0 kg/mm² to 14.1 kg/mm² there was little change in strain in the in-reactor test while the laboratory control test failed shortly after the increase in stress. The laboratory creep curve shown in fig. 5 at 220 °C and higher temperatures is a repeat test that was destressed after it reached 2% strain immediately following the increase in stress from 7 to 14 kg/mm² (going from part A to part B of fig. 5) and allowed to age-harden for 180 h at 200 °C before reloading to 14.1 kg/mm² at 220 °C. It would appear from the above that irradiation damage and strain-age-hardening have a similar effect on creep of zirconium and it is conceivable that some of the changes in the creep rate observed by Fillnow et al. [10]) may have been irradiation induced transients affecting the primary creep only.

There appeared to be a discrepancy between the in-reactor creep results obtained on 20% cold-worked Zircaloy-2 at Hanford and the early Chalk River tests. Holmes et al. [11]) reported the overall effect of neutron irradiation on the

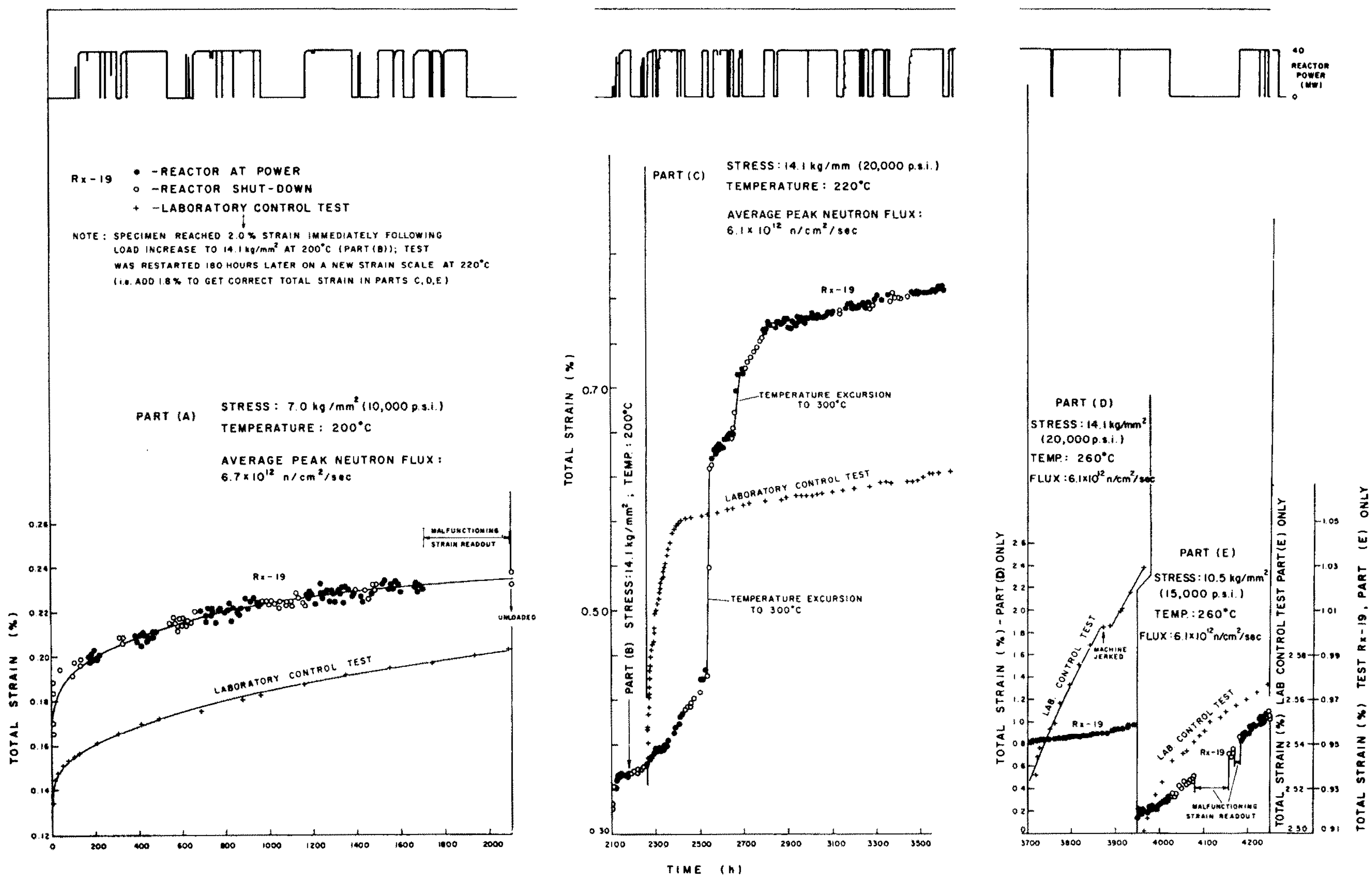

Fig. 5. In-reactor creep of annealed zirconium; stress: 7 to 14 kg/mm²; temperature: 200 to 300 °C; average peak neutron flux: 6 to 7×10¹² n/cm²·sec ($E>1$ MeV).

creep of 20% cold-worked Zircaloy-2 to be small, while at that time our measurements indicated a large effect of flux. Some of the Hanford tests exhibited increased creep rates following reactor shutdown while our tests either showed no change or a drop in creep rate. Whereas some of the differences might be attributed to differences in material preparation, i.e. the Chalk River material was given an additional stress relieving treatment of 72 h at 400 °C, much of the discrepancy was due to the different temperature ranges of testing. Nearly all early Chalk River results were obtained at 300 °C, while most of Hanford's data were above 350 °C. As discussed in section 5.3, there is a change in the temperature

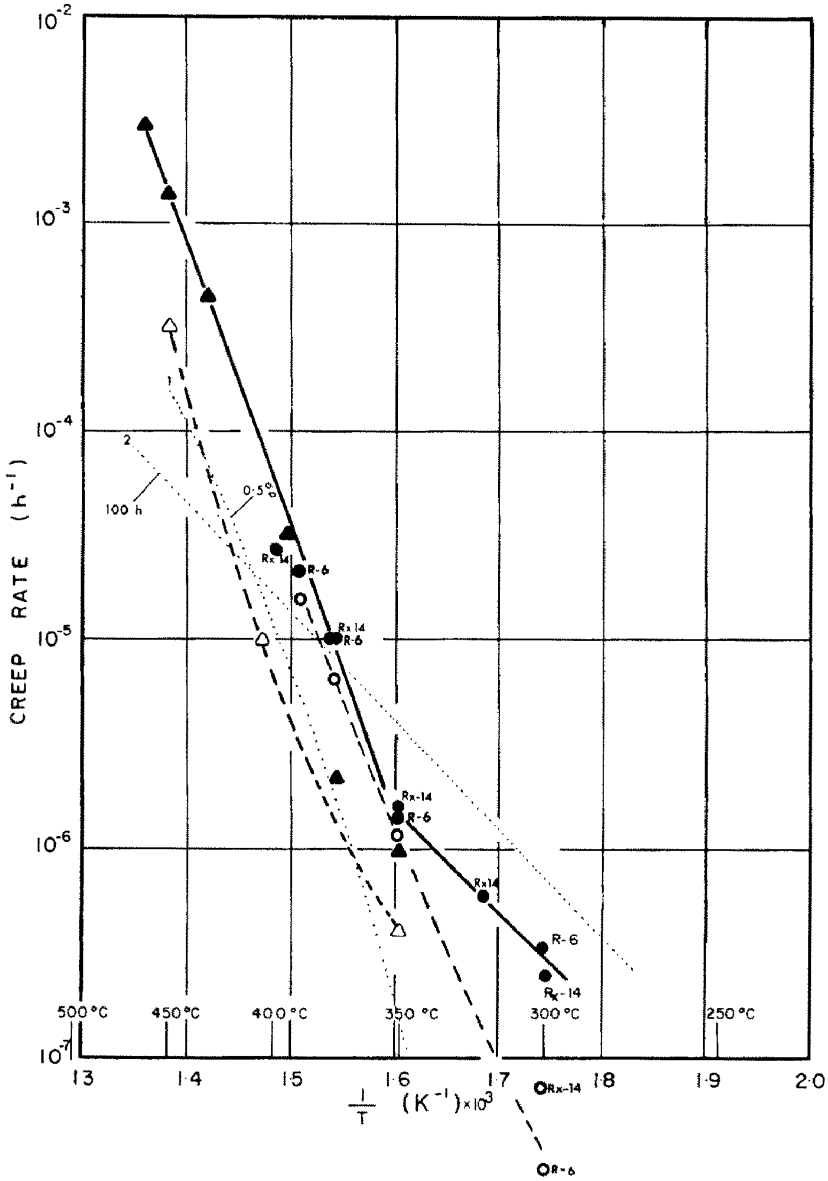

Fig. 6. Temperature dependence of in-reactor creep rate of cold-worked Zircaloy-2 at 14.1 kg/mm² stress (20 000 psi). ● AECL results in reactor; ○ AECL results in laboratory; ▲ Holmes et al. [11]) in reactor; △ Holmes et al. [11]) in laboratory; 1··· Ibrahim [16]) lab. data at constant strain of 0.5%; 2··· Ibrahim [16]) lab. data after constant time of 100 h.

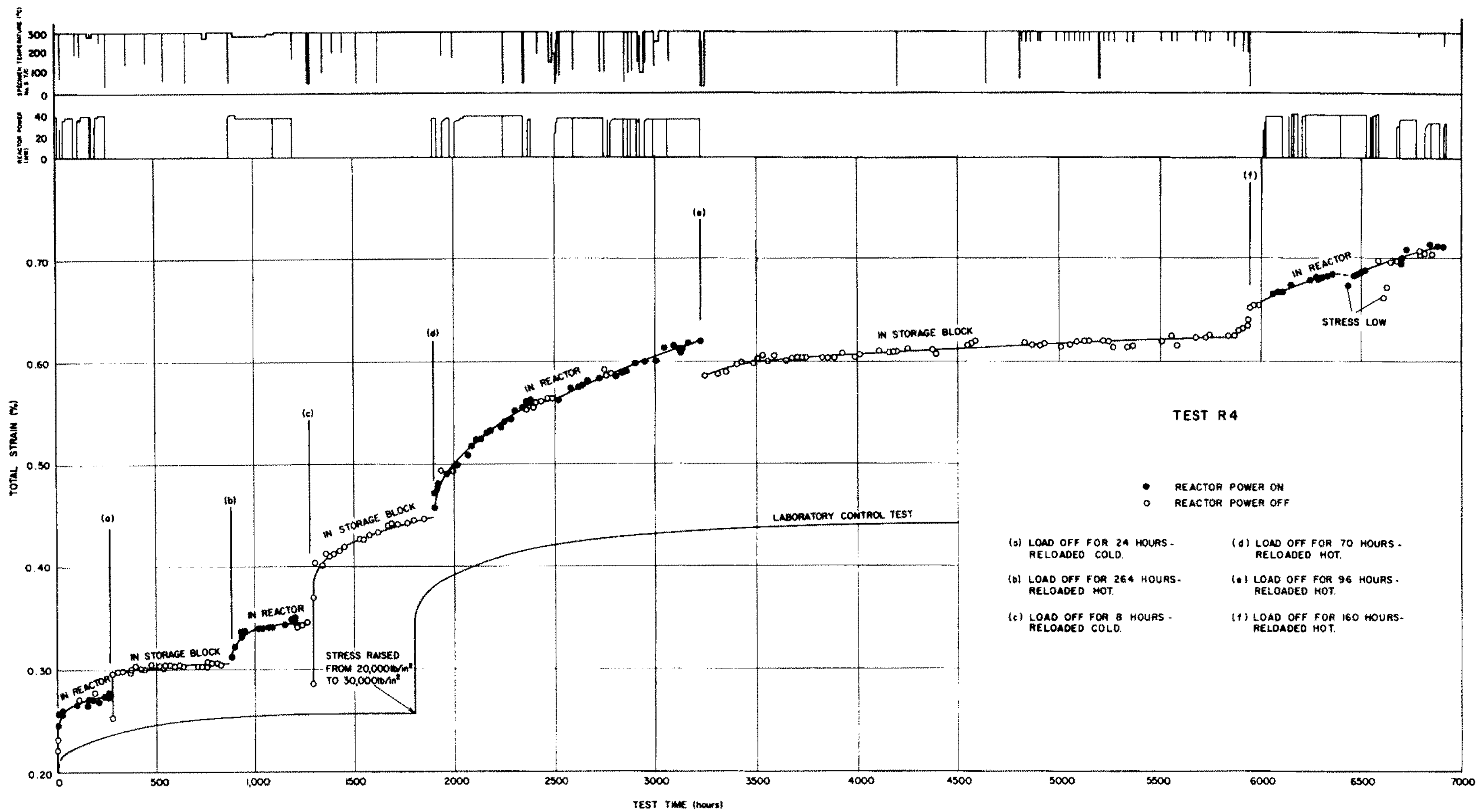

Fig. 7. In-reactor creep of Zircaloy-2 (cold-worked 20%, stress-relieved 72 h at 400 °C). Stress: 0–1280 h, 14.1 kg/mm^2 (20 000 psi); 1280–6920 h, 21.1 kg/mm^2 (30 000 psi). Temperature: 300 °C; peak neutron flux: 4–6$\times 10^{12}$ n/cm^2·sec (E>1 MeV).

dependence of in-reactor creep rate around 350 °C. Linear extrapolation to 300 °C from results obtained above 350 °C would therefore result in large discrepancies. Subsequent Chalk River tests at temperatures above 300 °C showed reasonable agreement with Hanford data (fig. 6), i.e. the irradiation effect on creep was small. On the other hand there was never any indication of an increase in the creep rate following reactor shutdown in our tests and therefore this observed difference in behaviour remains to be explained.

5.2. Flux dependence of the in-reactor creep rate

5.2.1. *Dependence on instantaneous flux*

The tests with the same specimen in different reactor positions (R-2, Rx-21) carried out to determine the relation between flux intensity and instantaneous creep rate were inconclusive because of scatter. There is some evidence (table 4, tests R-2 and R-4 at 21.1 kg/mm² stress) to suggest a linear flux dependence as reported by Ross-Ross and Hunt [12]) on cold-worked Zircaloy-2 and cold-worked Zr-2.5 wt % Nb pressure tubes. On the other hand the two tests Rx-13 and Ru-1 (fig. 2) that were carried out in widely different fluxes suggest a stronger flux dependence. Unfortunately test Ru-1 had numerous temperature cycles to 50 °C from 300 °C and the result is therefore suspect.

5.2.2. *Dose dependence*

The dependence of the in-reactor creep rate on the integrated flux has only been noticed at high stresses and high strain rates as discussed in section 5.1. The presence in large amounts of radiation induced obstacles is very effective in reducing the creep rate. For example, the creep rate of cold-worked Zircaloy-2 at 300 °C and 31.6 kg/mm² after pre-irradiation to 3×10^{20} n/cm² is lower, both in and out of flux, than for unirradiated material (test Rx-21, fig. 3). The effect seems to be small at low stresses and strain rates around 10^{-7}/h judging by the very similar creep rates in tests R-6 (no pre-irradiation) and Rx-14 (pre-irradiated to 3×10^{20} n/cm²), as well as the very nearly identical creep rates of test R-4 (section e to f, fig. 7) during prolonged out-reactor testing and its unirradiated laboratory control test. This agrees with other workers' creep, stress rupture and stress relaxation results [13–15]) on annealed and cold-worked Zircaloy-2 before and after irradiation.

5.3. Temperature dependence of the in-reactor creep rate

Figs. 6 and 8 show the temperature dependence of the in-reactor creep rate. Except for the test Rx-23 all creep rates above 300 °C were determined after relatively short testing time and were still slowly decreasing so the values are somewhat high. The temperature dependence of the in-reactor creep rate appears

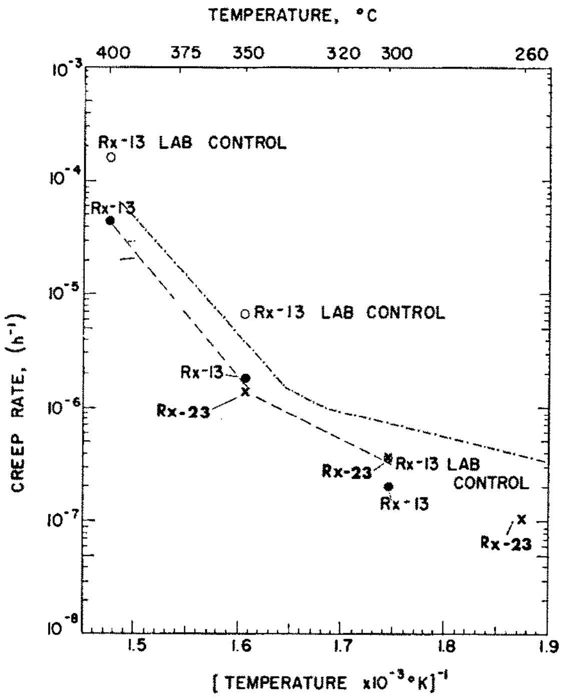

Fig. 8. Temperature dependence of in-reactor creep rate of Zircaloy-2 and Zr-2.5 wt % Nb.
— — — on 15–20 % c.w. Zr–2; stress = 14.1 kg/mm² (20 000 psi) — . — . — 15–20% c.w. Zr–2; stress = 21.1 kg/mm² (30 000 psi) ●○ on Zr–2.5 wt % Nb (quenched–30 % c.w.–aged) stress 16.2 kg/mm² × on Zr–2.5 wt % Nb (quenched–11 % c.w.–aged) stress 14.1 kg/mm² neutron flux = 5–9 × 10¹² n/cm² sec (E > 1 MeV).

to be similar for cold-worked Zircaloy-2 and quenched, cold-worked and aged Zr-2.5 wt %Nb. Around 350 °C there is a change in the slope of the inverse temperature vs creep rate plot. The Arrhenius plot is often used to derive a value for the activation energy of the rate controlling process. In a situation where there is only one rate controlling process, as for example during dislocation climb controlled creep at temperatures above half the melting point temperature of the metal, $\frac{1}{2}T_m$, the rate process is given by

$$\dot{\varepsilon} = A \exp\{-Q/(RT)\},$$

where

$\dot{\varepsilon}$ = creep rate,
Q = activation energy for creep,
R = gas constant,
T = temperature (°K),
A = structure factor.

The temperature coefficient which is given by the slope of the Arrhenius plot is often referred to as the activation energy for creep and, in the case of pure metals above $\frac{1}{2}T_m$ normally agrees closely with the activation energy for self-diffusion.

In the case of the in-reactor creep results the value of the temperature coefficient above 350 °C lies between 50 000 and 60 000 cal/mol for both Zircaloy-2 and Zr-2.5 wt % Nb in the stress range 14–21 kg/mm². Below 350 °C the value of the temperature coefficient is about 20 000 cal/mol at 14.1 kg/mm² and 10 000 cal/mol at 21.1 kg/mm².

Because of continuously decreasing creep rate the laboratory data do not in general yield a straight line on the Arrhenius type plot at temperatures below 450 °C, unless compared at the same creep strain or after identical test times. This type of analysis was carried out at 14.1 kg/mm² stress on creep results of cold-worked Zircaloy-2 reported by Ibrahim [16]). In the region in which experimental results were available approximately straight lines (fig. 6) were obtained on the Arrhenius plot for both constant strain and constant time conditions. For constant strain of 0.5% the slope gave a temperature coefficient of 67 000 ± 10 000 cal/mol in the temperature range 350–450 °C, while for a constant time condition of 100 h the temperature coefficient was 25 000 ± 5000 cal/mol in the temperature range 275–450 °C.

No particular significance is attached to these values, except as an empirical method for

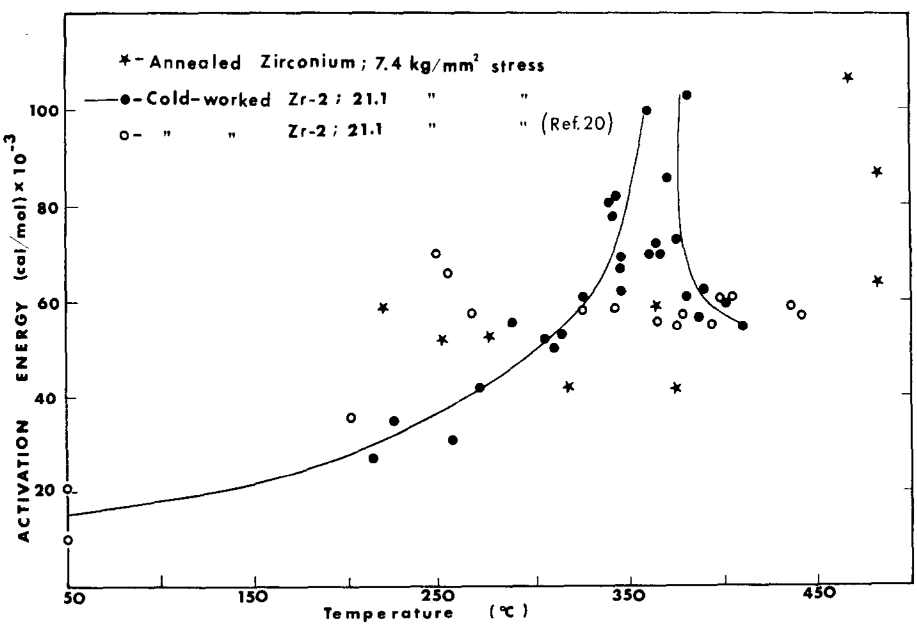

Fig. 9. Temperature dependence of activation energy for creep of zirconium and Zircaloy-2.

correlating experimental data, in spite of the proximity of some of the values of the temperature coefficient to the value of activation energy for diffusion of zirconium in Zr-1.3 wt % Sn alloy, reported by Lyashenko et al. [17]). In the review of diffusion processes in zirconium and its alloys Kidson [18]) questioned the reliability of Lyashenko's results and in the absence of other determinations considered the value of 45 500 cal/mol for self-diffusion of zirconium in α-zirconium obtained by Flubacher [19]) to be a more reliable value. Therefore, the claim that creep in Zircaloy-2 is diffusion controlled at temperatures as low as 250 °C which Holmes[11, 20]) makes on the basis of 58 500 cal/mol for the activation energy for creep does not seem to be justified.

The values of activation energy for creep for cold-worked and annealed Zircaloy-2 and annealed zirconium carried out in the laboratory by the Dorn's temperature cycle method (fig. 9) show considerable scatter and a marked peaking in the temperature range 250–450 °C. Similar peaking has been reported by Holmes [20]). This, combined with the knowledge that both annealed zirconium and annealed and cold-worked Zircaloy-2 exhibit continuously decreasing creep rates with time, as well as strain-age hardening characteristics [21, 22]), throws doubt on whether the use of activation energy values obtained from creep measurements is of any value for deducing creep mechanisms.

5.4. Stress dependence of the in-reactor creep rate

Fig. 10 shows the stress dependence of the

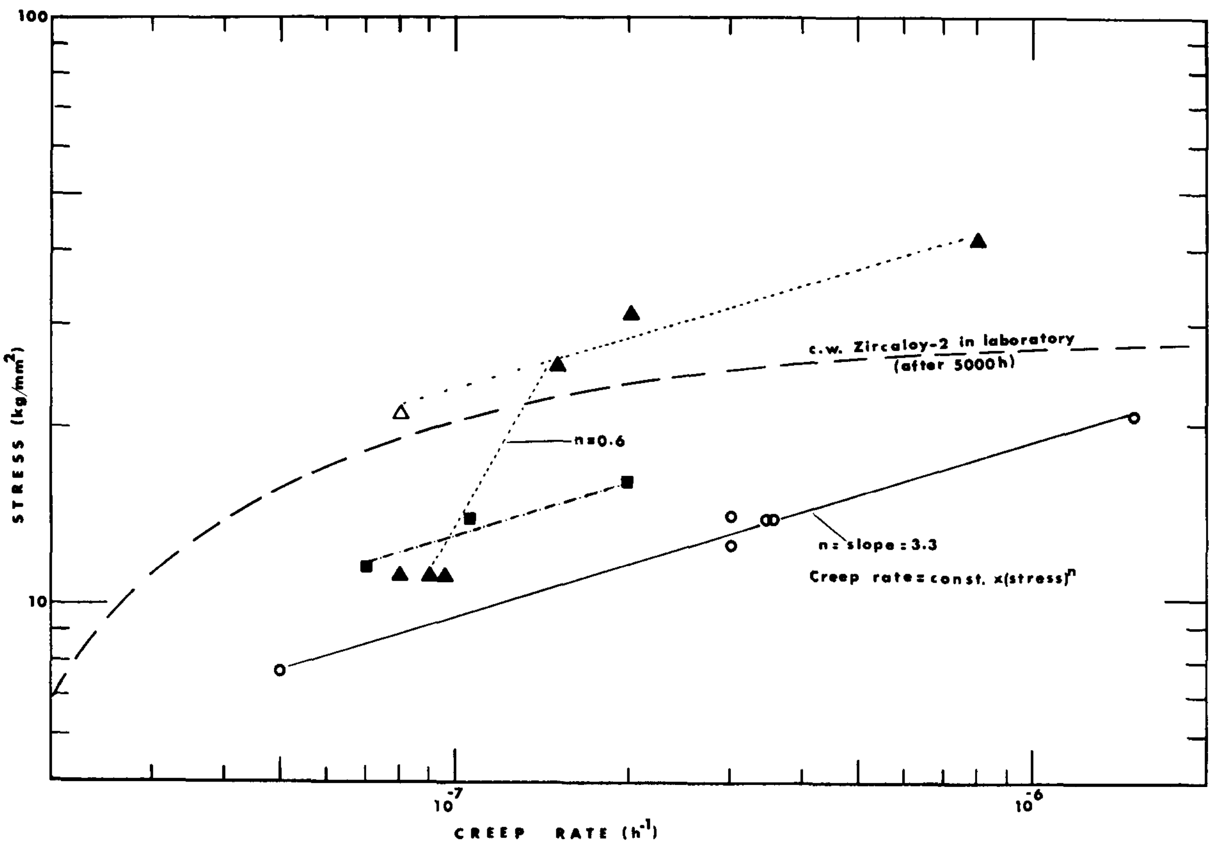

Fig. 10. Stress dependence of in-reactor creep rate at 300 °C; fast neutron flux: 5–7 $\times 10^{12}$ n/cm^2·sec (E>1 MeV).

——— ○ cold-worked Zircaloy-2 in reactor; —.—. ■ cold-worked Zr-2.5 wt % Nb in reactor; △ q. and aged Zr-2.5 wt % Nb in reactor; ------ ▲ q.-c.w.-aged Zr-2.5 wt % Nb in reactor.

in-reactor creep rate using a log-log scale. A creep rate proportional to the n^{th} power (n approximately equal to 3) of the stress could be fitted to the results of cold-worked Zr-2.5 wt % Nb and Zircaloy-2 as well as heat treated Zr-2.5 wt % Nb above 21.1 kg/mm² stress. In the lower stress range some of the heat treated Zr-2.5 wt % Nb results appear to be only slightly dependent on stress, while others follow cold-worked Zircaloy-2. Until additional evidence becomes available use of the available data from samples of different texture and degree of prior cold-work to define the stress exponent could be misleading.

The laboratory control tests do not yield a inear relation on the log-log plot and therefore cannot be expressed in terms of a simple power law.

5.5. Material dependence of the in-reactor creep rate

Fig. 11 shows the dependence of the in-reactor creep rate at 300 °C for all the samples measured to-date, normalized to a flux of 1×10^{13} n/cm² sec using a linear flux dependence. The general conclusions from samples cut in the longitudinal direction of pressure tubes and rods are:

a. Neutron flux increases the creep rate of cold-worked Zircaloy-2, cold-worked Zr-2.5 wt % Nb and heat treated Zr-2.5 wt % Nb.

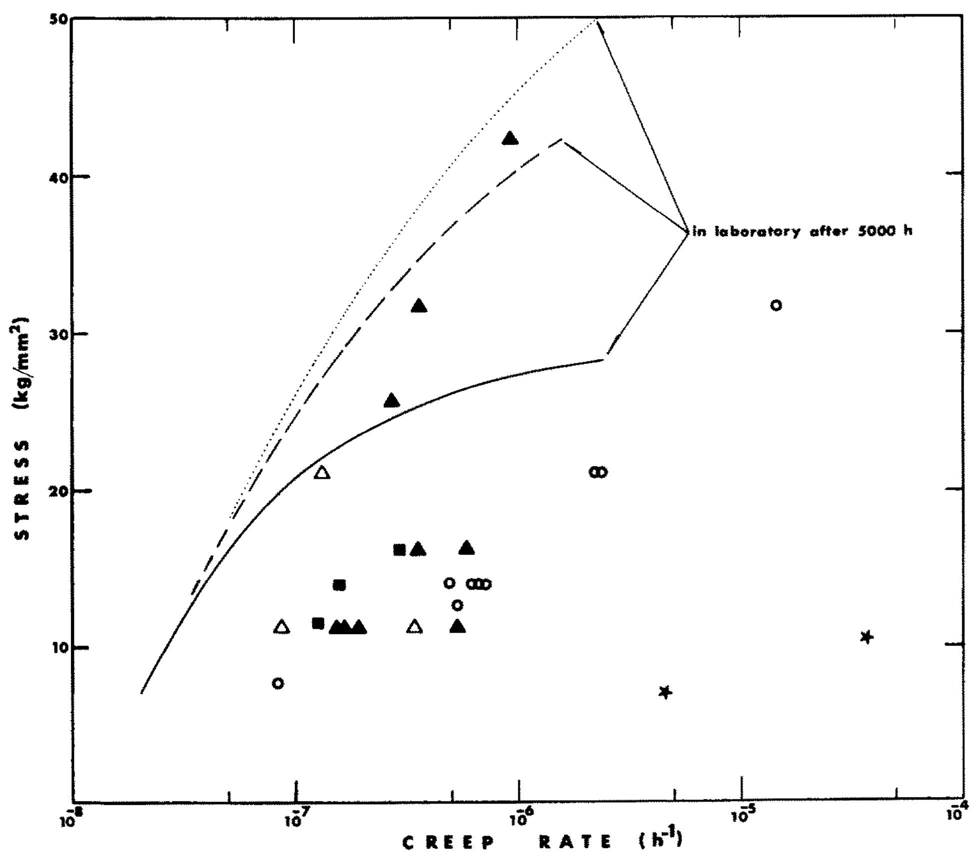

Fig. 11. In-reactor creep of zirconium alloys at 300 °C, normalized to a neutron flux of 1×10^{13} n/cm²·sec ($E > 1$ MeV) assuming linear flux dependence.
——— ○ c.w. Zircaloy-2; – – – ■ c.w. Zr-2.5 wt % Nb; -------- △ q. and aged Zr-2.5 wt % Nb; ▲ q.-c.w.-aged Zr-2.5 wt % Nb; ★ annealed Zr.

Comparison of the in-reactor and out-reactor tests indicates that the irradiation effect is highest in cold-worked Zircaloy-2 and lowest in heat treated Zr-2.5 wt % Nb.

b. Cold-worked Zr-2.5 wt % Nb has a lower in-reactor creep rate than cold-worked Zircaloy-2.

c. Zr-2.5 wt % Nb $(\alpha+\beta)$-quenched and aged 24 h at 500 °C has a much lower in-reactor creep rate than cold-worked Zircaloy-2 at stresses above 21.1 kg/mm^2. At lower stresses the information is not well defined, but appears to indicate a lower creep rate than for cold-worked Zircaloy-2.

d. Thirty percent cold-deformation between the quenching and aging of the heat treated Zr-Nb increases the in-reactor and laboratory creep rates by a large factor. The results with samples of lower intermediate cold-work indicated a lower in-reactor creep rate at stresses above 21.1 kg/mm^2 and a nearly stress independent in-reactor creep rate at lower stresses. The materials contained zones of recrystallization varying in extent with the amount of intermediate cold-work, as noted in table 3.

e. The irradiation effect on creep of annealed zirconium between 200–300 °C was not large enough to detect with existing instrumentation in tests of short duration.

5.6. Irradiation effect on start of tertiary creep

The laboratory creep results of Bell [23]) and Pankaskie [24]) indicated the onset of tertiary creep in cold-worked Zircaloy-2 to be around 1 to 2% strain. Both the tensile data on crept in-reactor specimens (see following section) and the information obtained from tests Rx-20 and Rx-21 indicate that there is no loss in ductility but possibly even some increase in uniform elongation and extension of the onset of tertiary to higher levels of strain than observed out-of-reactor, i.e. in both Rx-20 and Rx-21 the specimens reached approximately 2% strain without any indication of increasing creep rate.

5.7. Tensile properties of creep specimens

The room temperature tensile properties of in-reactor creep specimens are given in table 7. They show no significant loss in strength or ductility. Comparison with the tensile properties of unirradiated materials would appear to indicate reduction in total elongation after in-reactor creep, however, most of this apparent change resulted from the use of different dimensions for in-reactor specimens. The results in table 1 were obtained using 2.5 cm gauge length, 0.25–0.40 cm dia. tensile specimens, while those in table 7 were obtained on 5.0 cm gauge length 0.1–0.2 cm diameter creep specimens.

5.8. Mechanism of in-reactor creep

In an earlier paper [4]) various creep mechanisms were reviewed in terms of their possible contributions to the irradiation effect without reaching any definite conclusion. The situation has not changed appreciably since then, mainly because no significant progress has been made to define the mechanism controlling the out-reactor creep in zirconium alloys around 300 °C.

A suggestion was made by the author in an earlier paper [4]) that obstacles formed by neutron bombardment might reduce the in-reactor creep rate at temperatures where damage recovery rates were low, and increase the creep rate at temperatures where the damage begins to recover. However, tests at low stresses with cold-worked Zircaloy-2 and annealed zirconium in the temperature range 200–260 °C showed either no change in creep rate (annealed zirconium) or an increase (cold-worked Zircaloy-2) even though Howe's work [25]) indicated that there was no significant recovery at those temperatures.

There have been a number of other attempts [11, 14, 26–28]) to explain the irradiation effect on creep, but none of them are able to explain fully the results presented in this paper.

At least three mechanisms, irradiation growth, growth induced yielding and a dislocation climb process such as jog dragging appear possible.

TABLE 7

Room temperature tensile properties of in-reactor creep specimens (gauge length 5.08 cm).

Material	Code no. (table 1)	Creep test no.	Approx. integrated neutron dose (n/cm^2)	Gauge dia. (cm)	Prop. limit (kg/mm^2)	0.2% Y.S. (kg/mm^2)	UTS (kg/mm^2)	Uniform elongation (%)	Total elongation (%)	Reduction in area (%)
Zirconium	33	Rx-19	7×10^{19}	0.203	15.5	19.3	21.4	5.0	6.3	61
Zircaloy-2	9	R-2	4×10^{19}	0.099	43.6	58.4	62.6	1.1	2.0	–
Zircaloy-2	18	Rx-20	3.1×10^{20}	0.203	50.7	59.5	65.2	3.3	4.5	58
Zircaloy-2	25	Rx-14	3.7×10^{20}	0.203	49.0	51.5	61.2	6.5	7.7	34
Zircaloy-2	25	Rx-21	3.4×10^{20}	0.203	52.0	59.8	65.4	3.0	4.0	34
Zircaloy-2	42	Rx-15	4.3×10^{20}	0.203	60.7	66.2	67.5	4.4	6.0	32
Zr-2.5 wt % Nb	27	R-7	5×10^{19}	0.203	74.5	87.9	95.6	2.8	4.8	58
Zr-2.5 wt % Nb	34	R-11	5×10^{19}	0.203	58.3	65.4	80.8	6.5	8.0	23
Zr-2.5 wt % Nb	34	R-12	5×10^{19}	0.203	59.8	64.7	80.8	6.3	7.6	59
Zr-2.5 wt % Nb	39	Rx-18	3×10^{19}	0.203	82.1	92.3	92.3	6.5	2.5	64
Zr-2.5 wt % Nb	37	Rx-16	9×10^{19}	0.203	72.0	83.9	89.8	2.0	3.5	38
Zr-2.5 wt % Nb	51	Rx-24	3×10^{19}	0.203	43.1	50.3	52.0	1.6	2.9	36

The relative contribution of any one of them will depend on the alloy, its metallurgical condition and degree of prior cold-working. In cold-worked Zircaloy-2 jog dragging may be expected to be the controlling mechanism, while in annealed zirconium a transient dislocation climb of the type described by Hesketh [14]) may be followed by smaller but steady contribution due to anisotropic irradiation growth. A detailed discussion of mechanisms is presented by Piercy [9]) and Hesketh [29]).

6. Summary

At 300 °C increases in creep rate of up to an order of magnitude have been measured in a fast neutron flux of 1×10^{13} n/cm$^2\cdot$sec ($E > 1$ MeV) with specimens of cold-worked Zircaloy-2. The flux induced increases were smaller with cold-worked and heat treated Zr-2.5 wt % Nb and not measurable in annealed zirconium.

Prior irradiation has no significant effect on creep in the 10^{-7} h^{-1} range, but significantly reduces the creep rate in the 10^{-5} h^{-1} and higher ranges (the stress rupture range).

There is some information to indicate that the dependence of the in-reactor creep rate on flux intensity is linear, although at least one set of results has indicated the proportionality factor to be higher.

The stress dependence of the in-reactor creep rate appears to follow a power law in cold-worked Zr-2.5 wt % Nb, Zircaloy-2 and at high stresses (> 21 kg/mm^2) in heat treated Zr-2.5 wt % Nb alloys. The stress exponent is about 3. At lower stresses, the creep rates in some of the heat treated Zr-2.5 wt % Nb tests appeared to be only slightly dependent on stress.

The temperature dependence of the in-reactor creep rate changes around 350 °C in both cold-worked Zircaloy-2 and heat treated Zr-2.5 wt % Nb. Above 350 °C and 14 kg/mm^2 the dependence can be expressed by an activation energy of 50 000 to 60 000 cal/mol, while below 350 °C the activation energy value drops to about 20 000 cal/mol.

Irradiation seems to prolong the onset of tertiary creep in Zircaloy-2 to levels of strain beyond the 1 to 2% measured in the laboratory. The room temperature tensile properties of in-reactor creep specimens show no significant loss in strength or ductility.

No single mechanism will explain all the available in-reactor creep results. Irradiation growth, growth induced yielding as well as irradiation enhanced climb of jogs in dislocations must be considered as factors contributing to the observed increases in the in-reactor creep rate.

Acknowledgements

The author is grateful to Mr. L. C. Cushing, Mr. P. G. Anderson, Mr. I. Emmerton, Mr. F. C. Harris and Mr. M. Marciukaitis for carrying out the creep experiments, to Dr. C. D. Williams, Dr. G. R. Piercy and Dr. W. R. Thomas for many valuable discussions and to Mr. E. C. W. Perryman for continued support and encouragement.

References

1) B. A. Cheadle and C. E. Ells, Trans. AIME 233 (1965) 1044
2) B. A. Cheadle and C. E. Ells, Electrochem. Tech. 4 (1966) 329
3) V. Fidleris, AECL Report, to be published
4) V. Fidleris and C. D. Williams, Electrochem. Tech. 4 (1966) 258
5) C. D. Williams and R. W. Gilbert, J. Nucl. Mat. 18 (1966) 161 and paper presented at Tokyo Conf. Strength of metals and alloys (Sept. 1967)
6) C. D. Williams, V. Fidleris and L. G. Bell, Chalk River (Canada) Report, AECL-2615 (1967)
7) V. Fidleris, in USAEC Report TID-7697 (1964)
8) V. Fidleris, H. N. Isaac and H. P. Koehler, Chalk River (Canada) Report, AECL-2568 (1966)
9) G. R. Piercy, J. Nucl. Mat. (this issue, pp. 18–50)
10) F. E. Faris, 1st Intern. Conf. peaceful uses Atomic Energy, Geneva 7 (1955) 484
11) J. J. Holmes, J. A. Williams, D. H. Nyman and J. C. Tobin ASTM publ. STP 380 (1965) 385
12) P. A. Ross-Ross and C. E. L. Hunt, J. Nucl. Mat. (this issue, pp. 2–17)
13) N. E. Hinkle, ASTM publ. STP 341 (1963) 344
14) R. V. Hesketh, Phil. Mag. 8 (1963) 1321
15) W. R. Smalley, Westinghouse (USA) Report CVNA-159 (1962)

[16] E. F. Ibrahim, Chalk River (Canada) Report, AECL-2528 (1965)
[17] V. S. Lyashenko, V. N. Bykov and L. B. Pavlinov, Fiz. Metal. Metalloved 8 (1959) 362
[18] G. V. Kidson, Electrochem. Tech. 4 (1966) 193
[19] P. Flubacher, Würenlingen (Switzerland) Report, EIR-Bericht, Nr. 49 (1963)
[20] J. J. Holmes, J. Nucl. Mat. 13 (1964) 137
[21] R. L. Mehan and F. W. Wiesinger, Knolls (USA) Report, KAPL-2110 (1961)
[22] F. R. Shober, R. W. Getz, P. D. Schumaker, A. P. Young, M. F. Amateau and R. F. Dickerson, Battelle Memorial Institute (USA) Report, BMI-1616 (1963)
[23] L. G. Bell, Can. Metal. Quart. 2 (1963) 119
[24] P. J. Pankaskie, Hanford (USA) Report, HW-75267 (1962)
[25] L. M. Howe, Chalk River (Canada) Report, AECL-1484 (1962)
[26] R. V. Hesketh, Phil. Mag. 7 (1962) 1417
[27] R. G. Anderson and J. F. W. Bishop, Inst. Metals Monograph 27 (1962)
[28] V. S. Blackburn, Phil. Mag. 6 (1961) 503
[29] R.V. Hesketh, J. Nucl. Mat. (this issue, pp. 77–86)