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DEVELOPMENT
OF A
HIGH YIELD 90 Zr - 10 U ALLOY
MASSIVE HYDRIDE
(Title Unclassified)

AEC Research and Development Report

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ABSTRACT

Through addition of approximately 0.4 wt % carbon (as zirconium carbide) to a 90 wt % Zr - 10 wt % U alloy, a 100% yield of uncracked massively hydrided fuel elements of high hydrogen density ($N_H = 5.7$ to 6.6) was obtained. The marked increase in yield was attributed to the presence of a carbide phase, serving as a nucleant and grain-growth inhibitor during solidification and subsequent solid-state transformations that occur during the hydriding process. The resulting hydride structure was fine grained and equiaxed.

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Figure 1. Hydrided SNAP 2-10 Fuel Rod ($H/Zr \sim 1.80$),
Illustrating Typical Hydride Cracking

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Figure 2. Cross Section of 90 Zr - 10 U Alloy Hydride SNAP 2-10
Fuel Rod ($H/Zr \sim 1.80$), Illustrating Intergranular Fracture
Characteristic of Hydrided Material of this Composition

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I. INTRODUCTION

The preparation of crack-free, massive hydrides of zirconium-uranium alloys has long presented a seemingly insolvable problem. In the present application, cylindrical fuel rods (1-1/4 in. in diameter by 12-1/2 in. long, for SNAP 2-10, and 5/8 in. in diameter by 14 in. long, for SNAP-8) of 90 Zr - 10 U alloy were observed to crack during the hydriding cycle at a hydrogen concentration greater than $H/Zr = 1$,^{*} resulting in an extremely low uncracked massive hydride yield. A 20% yield was not considered unusually low. The fracture was invariably observed to be radial and intergranular in nature, usually extending the entire length of the rod.

A number of hypotheses were advanced to explain the cracking phenomenon, including the formation of oxides and other film barriers resulting in nonuniform hydrogen occlusion, nonuniform strain textures resulting from the extrusion process, and hydride temperature gradient effects resulting in nonuniform hydrogen diffusion. However, attempts to improve the yield, through minimizing these effects, met with little success. In addition, one salient fact, not consistent with any theory based on nonuniformity, was apparent, in that the cross-section macrostructure of the cracked rods was observed to be essentially uniform from end to end.[†] That is, if the rate or mechanism of hydrogen occlusion had been significantly different over the length of the rod, one would expect some degree of difference to be evident in the hydride macrostructures or growth textures. The rod shown in Figure 1 illustrates the type of failure generally observed. Figure 2 is a cross-section photomicrograph of a hydrided SNAP 2-10 fuel rod ($H/Zr \sim 1.8$), illustrating radial intergranular cracking characteristic of hydrided material of this composition. Note particularly the ring of large columnar grains extending radially inward from the rod surface. This large-grained radial columnar texture was observed to be the characteristic structure in all instances of hydride cracking.

Considering this structure, it is possible to postulate a mechanism whereby circumferential and longitudinal tensile stresses are generated in the outer surface of the rods, resulting in intergranular failure along the planes of weakness formed by the radial boundaries of these large columnar grains. To clarify

^{*} H/Zr is the atom ratio of hydrogen to zirconium in the hydride.

[†]Neglecting "end-effects," of course, where diffusion direction and overall diffusion rate are different.

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this point, one must consider physical changes occurring in the occluding matrix during hydriding. It has been proposed¹ that the columnar ring of coarse grains originates at the onset of the β (bcc) $\rightarrow$ δ (fcc) transformation, the δ being nucleated at the rod surface and growing radially inward as the transformation progresses, resulting in a preferentially oriented δ structure, aligned crystallographically so as to promote maximum heat transfer during the exothermic occlusion process. This transformation results not only in a decrease in bulk density,^{2,3} or conversely an increase in the specific volume of the transformed region, but also a drastic loss of ductility.^{3,4} It must then be clear that, once this ring of δ has formed, subsequent transformation and expansion of the inner core will place the outer δ ring in tension, ultimately resulting in rupture along the weak radial boundaries of the columnar δ grains.

Solution of the cracking problem, under this premise, lay in the elimination of the abnormal columnar growth tendency. The method was quite obvious; namely, inoculation with a foreign phase to serve as a nucleating agent and/or grain-growth inhibitor during the $\beta \rightarrow \delta$ transformation. However, it was not until the findings of Van Houten, in his work with growth inhibition in β -phase zirconium through additions of an insoluble foreign phase, that a promising nucleant was uncovered, namely carbon. Subsequent work by Van Houten with zirconium hydride,⁵ and by this investigator with 90 Zr - 10 U alloy hydride,⁶ yielded results beyond expectation. Through the addition of 0.4 wt % carbon, as zirconium carbide, an uncracked hydrided fuel rod yield of 100% was attained, for hydrogen concentrations as high as $H/Zr = 1.89$.

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II. MATERIALS

The melt stock used in extrusion billet preparation consisted of sponge zirconium, quarter-inch-cube sheared uranium stock, and -325 mesh zirconium carbide powder. Typical analyses of these materials are presented in Table I. The hydrogen used in the processing was prepurified bottle hydrogen, certified <10 ppm total impurity.

TABLE I
CHEMICAL COMPOSITION OF MELT STOCK
(ppm)

Element	Zirconium Sponge	Uranium	ZrC Powder
Ag		< 1	
Al	40	6	< 25
B	< 0.5	< 1	0.2
Ba		< 10	
Be		< 1	
C	< 10	200	11.39 wt %
Ca	< 20	< 50	
Cb			< 100
Cd	< 0.3	< 1	< 0.3
Cl	97		
Co	< 5	< 1	< 5
Cr	34	< 6	< 10
Cu	< 20	2	< 25
Fe	370	80	< 50
Hf	54		< 40
K		50	
Li	< 3	< 2	
Mg	<100	< 5	< 10
Mn	20	5	10
Mo	< 10	172	10
N	15		2300
Na	< 15	< 1	
Ni	< 10	< 30	< 10
O	686		1100
P	70	<100	
Pb	8		< 5
Si	35	275	< 40
Sn	< 50	< 10	< 10
Ta			< 200
Ti	< 20	< 4	< 20
V	< 10	< 10	30
W	< 40		< 25
Zn	< 25	< 10	< 50

7570-1873A

a. SNAP 8

7570-1873D

b. SNAP 2-10

Figure 3. Fuel Rod Loadings

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III. PROCEDURE

The 90 Zr - 10 U fuel alloy was prepared by the consumable electrode arc-melting method. Each billet was triple arc melted and subsequently extruded, at approximately 850°F, into 5/8-in. diameter rods for SNAP 8 and 1-1/4-in. diameter rods for SNAP 2-10. Details of the melting and forming operations are given elsewhere.⁷ Following extrusion, the rods were cut to length (14-1/2 in. for SNAP 8 and 12-3/4 in. for SNAP 2-10), and grit blasted to remove extrusion lubricant and scale. The rods were then centerless ground and degreased. They were then loaded in alumina refractory tubes of appropriate diameter, and capped with zirconium end plugs to eliminate end-effect radiant heat loss, and zirconium-turning "getter" plugs, and the loaded tubes were placed in boats. A load consisted of six SNAP 2-10 size rods, arranged in two concentric clusters of three, or eight SNAP 8 rods, in a single concentric cluster. Figures 3a and 3b illustrate the loading method used for SNAP 8 and SNAP 2-10, respectively. An end detail of the positioning of the loaded tubes in the boat is shown in Figures 4a (SNAP 8) and 4b (SNAP 2-10). The loaded boats were then placed in furnace retorts, capable of maintaining an isothermal profile of $\pm 10^\circ\text{F}$ over the entire length of the load. Views of the hydriding equipment, showing the retort loading end and the furnaces and programming controls, are shown in Figures 5a and 5b.

Subsequent to loading, the retorts were evacuated to 10^{-4} mm, and the charge programmed up to a temperature of 1650°F at 100°F/hr. Upon attaining thermal equilibrium, hydrogen* was admitted at such a rate as to attain an H/Zr ratio of 1.0 in the first 24 hr and H/Zr of 1.5 in the second 24 hr. The cycle to this point was maintained isothermal, pressure being established by the kinetics of the process. As hydrogen concentration requirements differ, for the two types of fuel rods (i. e., $N_H = 6.0 \pm 0.1$ for SNAP 8, $N_H = 6.5 \pm 0.1$ for SNAP 2-10), the final step in the hydriding cycle of the two materials differs appreciably. In the case of SNAP 8 processing, upon attaining an H/Zr of 1.5, the retort temperature was programmed down to 1370°F at 20°F/hr, while continuing to add hydrogen at the same rate as described in the previous step (i. e., H/Zr of 1 to H/Zr of 1.5 in 24 hr). Subsequent to establishing equilibrium

*Purified bottle hydrogen was used, certified 10 ppm maximum total impurity.

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7570-1873C

a. SNAP 8

b. SNAP 2-10

7570-1873B

Figure 4. End Views of Loaded Boats

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a. Showing Furnaces and Retorts From the Loading End

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b. Showing Furnaces and Program Control Panels

Figure 5. Hydriding Equipment

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at this temperature, the charge was programmed down the $H/Zr = 1.7$ isochore⁸ at $50F^{\circ}/hr$ to $1100^{\circ}F$, at which point the retort was isolated and furnace cooled. To attain the required SNAP 2-10 hydrogen concentration, upon attaining an H/Zr of 1.5 as described previously, the retort pressure was increased to 6000 mm by maintaining the previous hydrogen flow rate. After establishing equilibrium at this pressure and $1650^{\circ}F$, the retort temperature was programmed down at $10F^{\circ}/hr$ to $1400^{\circ}F$, and the system again allowed to equilibrate* while maintaining 6000 mm hydrogen pressure. The charge was then programmed down to $1200^{\circ}F$, at the same cooling rate, and a final equilibrium* was established at this temperature and 6000 mm. The retort was then isolated, the power shut off, and the charge furnace cooled. A detailed procedure of the hydriding process is given in References 9, 10, and 11.

*It is probable that a true equilibrium was not established at these lower temperatures, since occlusion kinetics become extremely sluggish at lower temperatures and excessive hydrogen concentrations (i.e., H/Zr approaching 2). Perhaps more correct phraseology, in this sense, would be to refer to the "equilibrium" as hydrogen saturation within a specified time period and at the temperature and pressure considered.

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IV. RESULTS AND DISCUSSION

The data regarding yield and hydrogen concentration of hydrided SNAP 8 and SNAP 2-10 fuel rods are presented in Tables II and III, respectively. The data in Table II are a compilation of results obtained with 24 SNAP 8 fuel rods, representing three separate and consecutive furnace loadings. As may be seen from the table, the rods were all crack free, and had a maximum variance in gross hydrogen concentration of $H/Zr = 1.65 \pm 0.04$. Similarly, the data in Table III were obtained from 24 SNAP 2-10 fuel rods, representing four separate and consecutive furnace loadings. Again the rods were all crack free, the variance in gross hydrogen concentration being $H/Zr = 1.82 \pm 0.03$.

Regarding the grain-refining effect of carbon on the Zr - 10 wt % U alloy, macrostructures of both modified and unmodified triple-arc-melted alloy are shown in Figures 6 and 7, respectively. The extreme grain refinement is apparent in the two photomicrographs. Also significant is the isotropic growth tendency displayed by the carbon-modified material, evident in the longitudinal and transverse sections of Figure 6. Fracture surfaces of carbon-modified and unmodified Zr - 10 wt % U alloy hydride are shown in Figures 8 and 9. The grain-refining effect of carbon on the hydride is readily apparent. The microstructure of carbon-modified alloy hydride appears in Figure 10. The striking uniformity of this structure, as compared to that of the alloy hydride sans carbon (Figure 2), is noteworthy. Regarding this structure, the banded matrix is composed of epsilon (fct) hydride, in which are dispersed a uranium-rich phase (fine spheroidal particles) and a carbon-rich phase* (larger spheroids).

*Probably ZrC, or a complex thereof, with uranium. Positive identification of this phase is not available, at this time.

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TABLE II
HYDRIDE YIELD - SNAP 8 FUEL RODS

Rod Number	Run Number	Rod Condition	H (wt %)	C (wt %)	H/Zr	N _H
559A-5	5-49	Sound - uncracked	1.62	0.51	1.66	5.89
559B-2			1.65	0.42	1.69	6.00
559B-3			1.64	0.43	1.68	5.96
559B-4			1.65	0.46	1.69	6.00
560D-1			1.63	0.42	1.67	5.93
560D-2			1.61	0.42	1.65	5.85
560D-3			1.61	0.43	1.65	5.85
560D-4			1.65	0.44	1.69	6.00
559C-1	6-47	Sound - uncracked	1.59	0.52	1.62	5.78
559C-2			1.61	0.52	1.65	5.85
559C-3			1.60	0.48	1.63	5.82
559C-4			1.60	0.50	1.63	5.82
559D-1			1.58	0.46	1.61	5.74
559D-2			1.60	0.45	1.63	5.82
559D-3			1.62	0.49	1.66	5.89
559D-4			1.64	0.50	1.68	5.96
561A-1	2-38	Sound - uncracked	1.61	0.51	1.65	5.85
561A-2			1.61	0.52	1.65	5.85
561A-3			1.60	0.51	1.63	5.82
561A-4			1.59	0.50	1.62	5.78
562B-1			1.60	0.49	1.63	5.82
562B-2			1.61	0.50	1.65	5.85
562B-3			1.60	0.51	1.63	5.82
562B-4			1.59	0.52	1.62	5.78

TABLE III
HYDRIDE YIELD - SNAP 2-10 FUEL RODS

Rod Number	Run Number	Rod Condition	H (wt %)	C (wt %)	H/Zr	N _H
201-6	1-24	Sound - uncracked	1.77	0.32	1.81	6.44
200-2			1.81	0.27	1.85	6.58
201-5			1.78	0.29	1.82	6.47
203-6			1.80	0.41	1.84	6.54
200-6			1.80	0.31	1.84	6.54
200-3			1.78	0.30	1.82	6.47
204-1	1-25	Sound - uncracked	1.75	0.28	1.79	6.36
203-2			1.76	0.35	1.80	6.40
203-5			1.78	0.41	1.82	6.47
204-2			1.77	0.29	1.81	6.44
202-6			1.80	0.34	1.84	6.54
203-4			1.76	0.38	1.80	6.40
207-1	3-41	Sound - uncracked	1.78	0.42	1.82	6.47
204-3			1.76	0.28	1.80	6.40
207-2			1.77	0.44	1.81	6.44
204-4			1.80	0.27	1.84	6.54
204-5			1.81	0.28	1.85	6.58
207-3			1.75	0.46	1.79	6.36
205-5	4-41	Sound - uncracked	1.75	0.63	1.79	6.36
206-5			1.78	0.34	1.82	6.47
205-6			1.79	0.52	1.83	6.51
206-3			1.79	0.30	1.83	6.51
206-4			1.75	0.36	1.79	6.36
205-4			1.79	0.62	1.83	6.51

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Figure 6. Triple-Arc-Melted Carbon-Modified 90 Zr - 10 U Billet Before Extrusion, Illustrating Grain-Refining Effect of Carbon (Compare this structure with that of unmodified alloy in Figure 7)

Figure 7. Triple-Arc-Melted Unmodified 90 Zr - 10 U Billet Before Extrusion

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Figure 8. Fracture Surface of Carbon-Modified 90 Zr - 10 U Alloy Hydride ($H/Zr \sim 1.7$), Illustrating Fine Uniform Fracture Characteristic of this Fine-Grained Structure (Compare this fracture with that of unmodified fuel material in Figure 9)

Figure 9. Fracture Surface of Unmodified 90 Zr - 10 U Alloy Hydride ($H/Zr \sim 1.7$), Illustrating Coarse Irregular Fracture Characteristic of This Material

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a. 500X

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b. 1000X

3076-3

Figure 10. Microstructure of Carbon-Modified 90 Zr - 10 U Alloy Hydride ($H/Zr \sim 1.8$) [This structure is composed of an epsilon (fct) matrix containing dispersions of a uranium-rich phase (fine spheroidal particles) and a carbon-rich phase (larger irregular spheroids).]

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V. CONCLUSIONS

Through a developmental program, consisting of a SNAP fuel alloy modification study and hydriding cycle evaluation, a yield of 100% crack-free massive hydrides, of hydrogen density $N_H = 5.74$ to 6.58 , was obtained. The addition of ~ 0.4 wt % carbon (as zirconium carbide) was found not only to inhibit grain-growth tendencies in Zr - 10 wt % U, during transformation from liquid to solid state and subsequent solid-state transformation occurring during the hydride process, but to restrict development of orientation textures on hydriding, resulting in equiaxed structures. Through minimization of preferred orientation, one may anticipate near-isotropic heat transfer, thermal expansion, and diffusivity characteristics for the modified alloy. However, these improvements were gained at the expense of increased hydrogen dissociation pressure. A complete evaluation of the magnitude of pressure increase is not available, at this time. However, preliminary data indicate carbon ties up an equal atom percentage of zirconium, thus decreasing the atom fraction of zirconium that may enter into the hydride reaction. This results in an increased hydrogen dissociation pressure for the carbon-modified fuel alloy, relative to unmodified fuel.

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