

663575

23

DP-1138

AEC RESEARCH AND DEVELOPMENT REPORT

DIRECTIONAL STRAIN AND HYDRIDE ORIENTATION IN ZIRCALOY

R. P. MARSHALL

SRL
RECORD COPY



Savannah River Laboratory

Aiken, South Carolina

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Printed in the United States of America

Available from

Clearinghouse for Federal Scientific and Technical Information

National Bureau of Standards, U. S. Department of Commerce

Springfield, Virginia 22151

Price: Printed Copy \$3.00; Microfiche \$0.65

663575
DP-1138

Metals, Ceramics, and Materials
(TID-4500)

DIRECTIONAL STRAIN AND HYDRIDE ORIENTATION IN ZIRCALOY

by

ROBERT P. MARSHALL

Approved by

P. H. PERMAR, RESEARCH MANAGER
NUCLEAR MATERIALS DIVISION

OCTOBER 1968

**E. I. DU PONT DE NEMOURS & COMPANY
SAVANNAH RIVER LABORATORY
AIKEN, S. C. 29801**

*CONTRACT AT(07-2)-1 WITH THE
UNITED STATES ATOMIC ENERGY COMMISSION*

ABSTRACT

A refined "Directional Strain Parameter" (D ϵ P-II) was developed to explain the hydride orientation in seamless Zircaloy tubing on the basis of laboratory and commercial fabrication histories. The directional strain (or fabrication) history, expressed as D ϵ P-II, explained 93% of the observed variations in hydride orientation in the tubing. The grain size of the tubing had a marginal effect on the hydride orientation, and the location of the hydride platelets -- transgranular or intergranular -- was not significant in controlling the orientation.

CONTENTS

	<u>Page</u>
LIST OF TABLES AND FIGURES	4
INTRODUCTION	5
SUMMARY	6
DISCUSSION	7
BACKGROUND	7
Hydride Orientation and Fabrication Strains	7
EXPERIMENTAL PROGRAM	7
Evaluation of D ϵ P by Laboratory Tests	10
Evaluation of D ϵ P by Commercial Fabrication Tests	12
RESULTS	18
Refinement of D ϵ P	18
Effect of Grain Size	19
Effect of Hydride Location	20
Statistical Analysis	21
Significance of Parameters	22
Significance of Zirconium Grain Size and Hydride Location	22
Definition of D ϵ P-II	24
Tests of D ϵ P-II	25
CONCLUSIONS	26
REFERENCES	27

LIST OF TABLES AND FIGURES

<u>Table</u>		<u>Page</u>
I	Fabrication Data for Laboratory-Swaged Zircaloy-2 Tubing	10
II	Chemical Analysis of Commercial Zircaloy-2 Tubes .	12
III	Data for Series-1 Commercial Zircaloy-2 Tubing . .	14
IV	Data for Series-2 Commercial Zircaloy-2 Tubing . .	14
V	Data Sets for Comparison of $\Delta\bar{D}$ and ΔW	19
VI	Orientation and Location of Hydride Platelets in Commercial Zircaloy Tubing	20
VII	Statistical Data from Analysis of Hydride Orientations in Commercial Tubing	23
 <u>Figure</u>		
1	Frequency Distributions of Hydride Platelet Orientations in Mandrel-Drawn and Tube-Reduced Zircaloy-2 Tubing	9
2	Frequency Distributions of Hydride Platelet Orientations in Swaged Zircaloy-2 Tubing	11
3	Fabrication Sequences for Series-1 Commercial Zircaloy-2 Tubing	13
4	Fabrication Sequences for Series-2 Commercial Zircaloy-2 Tubing	13
5	Hydride Orientations in Series-1 Zircaloy-2 Tubing.	15
6	Hydride Orientations in Series-2 Zircaloy-2 Tubing.	16
7	Hydride Orientation as Related to Total Strain History	18

INTRODUCTION

Zirconium alloys normally absorb corrosion-produced hydrogen during reactor service. This absorption causes hydride platelets to precipitate in the alloy when the solubility limit of hydrogen is exceeded.

The orientation of the hydride platelets in zirconium alloys has been studied extensively since the discovery of variable hydride orientations¹ and the effect of hydride orientation on ductility at ambient temperature. At the operating temperatures of many nuclear applications (150-200°C), Zircaloy is not susceptible to hydride embrittlement and thus retains its tensile properties.³ However, the embrittlement persists at ambient temperatures, and both reactors and fuel elements periodically cycled through this temperature range. Therefore, the factors that affect the orientation of hydride platelets in Zircaloy are important from an industrial standpoint.

The factors controlling the "natural" hydride orientation (hydride platelets formed with no externally applied stresses) in commercial Zircaloy tubing have been investigated. Kearns and Woods⁴ correlated, in the ring sections from small-diameter sheath tubing, the natural orientation of individual hydride platelets with the macroscopic crystallographic texture by assuming (0002) hydride plane habit. Hindle and Slattery⁵ found that the general orientation of hydride clusters in pressure tubing was not predominately related to texture, but appeared to be primarily a function of the comparative amounts of "pinch and sink" (reductions in wall and diameter) in the last fabrication process.

Studies at the Savannah River Laboratory (SRL)^{6,7} confirmed the generalized results of Hindle and Slattery, and compared the preferred orientations of individual hydride platelets to crystal-line textures and the thermomechanical fabrication history for large-diameter sheath tubing. Hydride orientation did not appear to be controlled by the macroscopic texture, but could be correlated with the strain history of the tubing.

A Directional Strain Parameter, D ϵ P (D ϵ P = Δ average diameter - Δ wall thickness) was developed to express the biaxial fabrication strains. These studies⁴⁻⁷ were post-fabrication investigations in which the hydride orientations after fabrication were compared to some property or to the history of the tubing.

This report describes three series of fabrication tests in which the natural hydride orientations were determined after each major process step. The purpose of this investigation was to test the directional strain concept and to refine the Directional Strain Parameter. The first fabrication test used a four-die rotary swager at the Savannah River Laboratory. The other two tests involved the full-scale fabrication of Zircaloy tubing by a commercial fabricator.

SUMMARY

The three fabrication tests confirmed the relationship between directional strain history and preferred platelet orientation. Reductions in tube diameter and wall thickness were the major factors that controlled platelet orientation.

Multivariable statistical analyses refined the Directional Strain Parameter and defined D ϵ P-II as [Δ average diameter - 0.475 Δ wall thickness]. D ϵ P-II explained 93% of the variation in platelet orientation, and was statistically significant much beyond the 10^{-3} level.

Zirconium grain size, which varied from 0.005 to 0.029 mm, was only marginally significant (0.04 level). The percent of platelets precipitated within the grains (58 to 89%) had no statistical significance on effect of platelet orientation.

DISCUSSION

BACKGROUND

HYDRIDE ORIENTATIONS AND FABRICATION STRAINS

The development of the Directional Strain Parameter⁷ (D ϵ P) was based on the phenomenon of "strain orientation" of hydride platelets. Early studies^{1,8} showed that plastic strain caused hydride platelets to precipitate more parallel to the direction of tensile strains, and that the effects were opposite for contractile strains. This phenomenon was used to analyze the fabrication history and the hydride orientation properties of the several types of tubing. The post-fabrication results were rationalized in two ways.

In an *additive* or *total fabrication* concept, each successive fabrication process added to or modified the prior hydride orientations. Successive fabrication processes with the same D ϵ P would further modify the hydride orientations.

In an *equilibrium* concept, the D ϵ P described a combination of strains that had associated with it a specific hydride orientation. Successive fabrication processes with the same D ϵ P would produce a characteristic hydride orientation that would not change during fabrication as long as the D ϵ P of each process remained constant. In the first study,⁷ the equilibrium concept appeared to be more tenable.

EXPERIMENTAL PROGRAM

Experiments were designed to: (1) test the correlation of directional strain with hydride orientation through successive fabrication processes, (2) determine the magnitude of the influence of different fabrication processes on the D ϵ P-hydride orientation relationship, (3) investigate possible differences due to the number of processing steps taken to arrive at a final tubing size, and (4) refine D ϵ P if it was found to be a significant factor in the control of hydride orientation.

Hydride orientation and fabrication strains were determined from samples cut from the tubes after each step in the fabrication process. At least twelve measurements each of outer diameter and wall thickness were used in calculating fabrication strains.

To analyze hydride orientation, one-inch-square samples were hydrogenated to 60 ppm at 400°C. The samples were cooled in the furnace (250°C/hr at 250-300°C) and prepared metallographically for observation of the tranverse planes of the tubing.

The orientation of *individual* hydride platelets was measured with a special eyepiece and electronic equipment.⁶ An "individual platelet" was defined as each straight segment of hydride phase. By this definition, a continuous volume of hydride phase made up of four straight segments was counted and analyzed as four platelets. The location of the platelet -- within a grain or at grain boundaries -- was also noted.

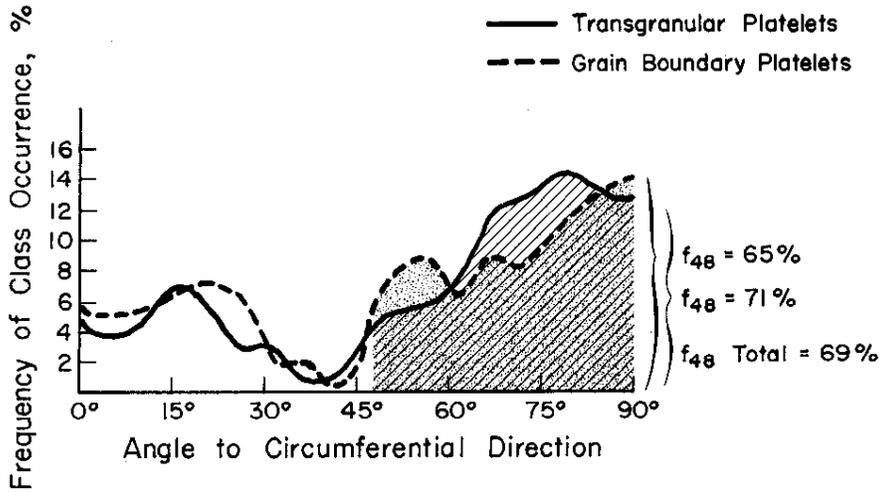
Orientation data were separated by computer into a frequency distribution of fifteen classes for each platelet location. Typical frequency distributions of the orientation of transgranular and grain boundary hydride platelets in two materials are shown in Figure 1. The development of the frequency distribution curves is described in more detail in Reference 7.

The preference of hydride platelets for a particular direction in the tubing was indexed by f_{48} , defined as the percent of platelets oriented between 48° and 90° to the reference direction (usually taken as the circumferential direction).

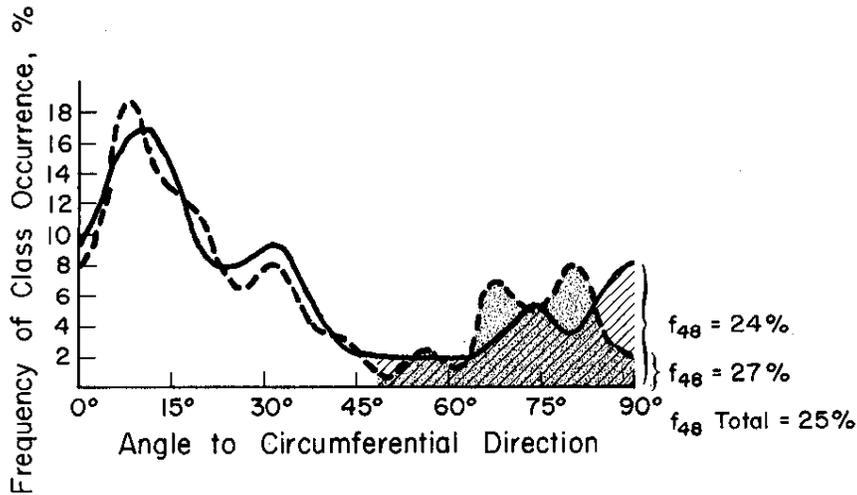
Hydride counting techniques and experimental variables were selected so that the data would represent a significant volume of each tube specimen. The data were analyzed to obtain a statistically significant hydride distribution profile.

The hydrogen concentration was set at 60 ppm to avoid counting excessive numbers of hydride platelets. A relatively fast cooling

rate from the hydrogenation treatment caused small, hydride platelets rather than the complex hydride clusters to precipitate.



a. Mandrel-Drawn Tubing, 22b (Series - 2)



b. Tube-Reduced Tubing, 15a (Series - 1)

FIG. 1 FREQUENCY DISTRIBUTIONS OF HYDRIDE PLATELET ORIENTATIONS IN MANDREL-DRAWN AND TUBE-REDUCED ZIRCALOY-2 TUBING (Transverse Tubing Plane)

EVALUATION OF DeP BY LABORATORY TESTS

The ability to control hydride orientation by control of directional strain was tested by a swaging experiment designed to measure the effect of small, carefully planned fabrication strains on the predominant hydride orientation. Fabrication sequences were specifically selected to change the hydride orientation from circumferential to radial and back to circumferential. A section of tubing with a hydride orientation (f_{48}) of 38% (Sample F in Reference 7) was given three passes in a four-die, rotary swager in which the diameter was substantially reduced with little change in wall thickness. These "sinking" passes were followed by two swaging passes that substantially reduced wall thickness with very small changes in diameter.

The tubing was annealed for 1/2 hour at 750°C and measured after each operation. Samples for hydride orientation measurements were taken after the first, third, fourth, and fifth anneals. The fabrication data are given in Table I.

TABLE I
Fabrication Data for Laboratory-Swaged Zircaloy-2 Tubing

Sample	Outer Diameter, in.	Wall Thickness W, in.	Average Diameter \bar{D} , in.	$\Delta\bar{D}$, %	ΔW , %	DeP [$\Delta\bar{D} - \Delta W$], %
F	2.530	0.037	2.493	-	-	-
S1	2.367	0.038	2.329	-6.6	+2.7	-9
	Anneal 30 min at 750°C					
S2	2.276	0.037	2.239	-3.9	-2.6	-1
	Anneal 30 min at 650°C					
S3	2.182	0.040	2.142	-4.3	+8.1	-12
	Anneal 30 min at 750°C					
S4	2.161	0.038	2.122	-0.9	-5.0	+4
	Anneal 30 min at 750°C					
S5	2.153	0.036	2.116	-0.3	-5.3	+5
	Anneal 30 min at 750°C					

The frequency distribution graphs of the hydride orientations of each sample are given in Figure 2. The three "sinking" swaging processes, S1-S2-S3, increased the fraction of radial hydride

platelets (f_{48}) from 38 to 73% with small amounts of deformation, i.e., changes in cross sectional area, less than 5% per pass. The dimensional changes produced in these passes were described by negative directional strain parameters (D ϵ P), S1 and S3, in Figure 2. During the next two swaging sequences, the radial platelets decreased from 73 to 61 percent. These sequences were "wall thinning" passes and were described by positive D ϵ P's (S4 and S5 in Figure 2). Qualitatively, these results demonstrated that the natural hydride orientations in Tube F could be controlled by small but directed fabrication strains. Further, the agreement between the sign of the Directional Strain Parameters for each swaging sequence and the direction of the resultant change in hydride orientation indicated the D ϵ P was adequately defined to justify a full-scale test with commercial seamless tubing.

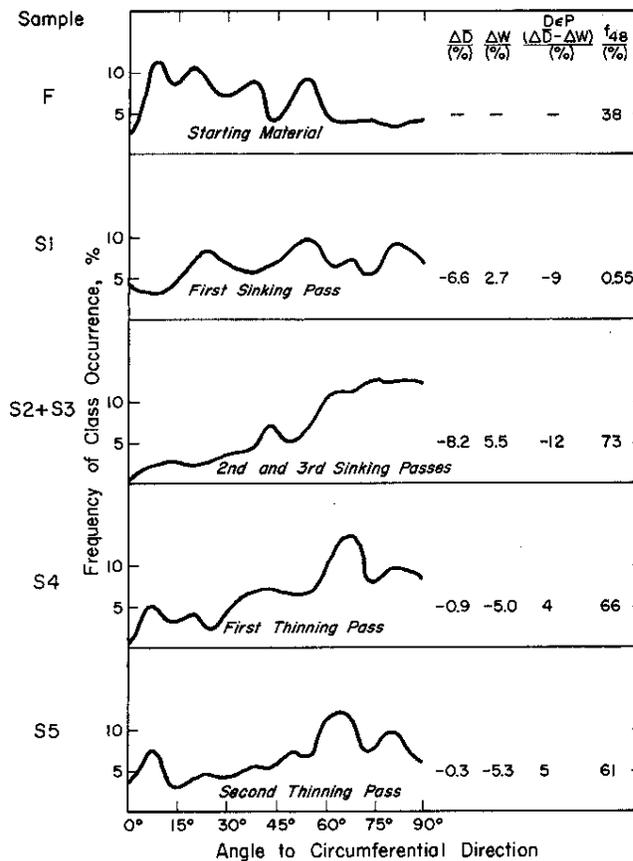


FIG. 2 FREQUENCY DISTRIBUTIONS OF HYDRIDE PLATELET ORIENTATIONS IN SWAGED ZIRCALOY-2 TUBING

EVALUATION OF DεP BY COMMERCIAL FABRICATION TESTS

Commercial fabrication of the test pieces began with 1400-1500°F extrusion of two Zircaloy-2 shells. This extrusion caused reductions of diameter and wall thickness of 50 and 90%, respectively. Chemical analysis of the two shells are given in Table II. The fabrication sequence for Shell 1 was designed to make a base tube with a preference for formation of circumferential hydrides, and to allow determinations of how quickly various fabrication sequences would change this preferred hydride orientation. Figure 3 summarizes the fabrication sequence. Shell 2 was processed to yield a base tube with a preference for formation of radial hydrides, and to allow studies of how effectively various fabrication steps changed the hydride orientations. Figure 4 summarizes the fabrication sequence. The tubes were annealed 2 hours at 700°C after each fabrication sequence; samples for hydride orientation measurements were taken before and after each anneal. The hydride orientation data always refers to annealed tubing unless specifically noted. The fabrication and hydride orientation data are given in Tables III and IV.

TABLE II
Chemical Analyses of Commercial Zircaloy-2 Tubes

<u>Element</u>	<u>Series-1</u>	<u>Series-2</u>
		wt %
Cr	0.11	0.10
Fe	0.15	0.14
Ni	0.05	0.05
Sn	1.39	1.46
		ppm
Al	45	37
C	110	100
Cb	<100	<100
Hf	72	61
N	43	51
O	1020	1180
Si	55	64
Ta	<200	<200
Ti	22	36
W	<25	<25
B, Ca, Cd, Cu, Mg, Mn, Mo, Pb, U, V, Co	<15	<15
Zr	Bal.	Bal.

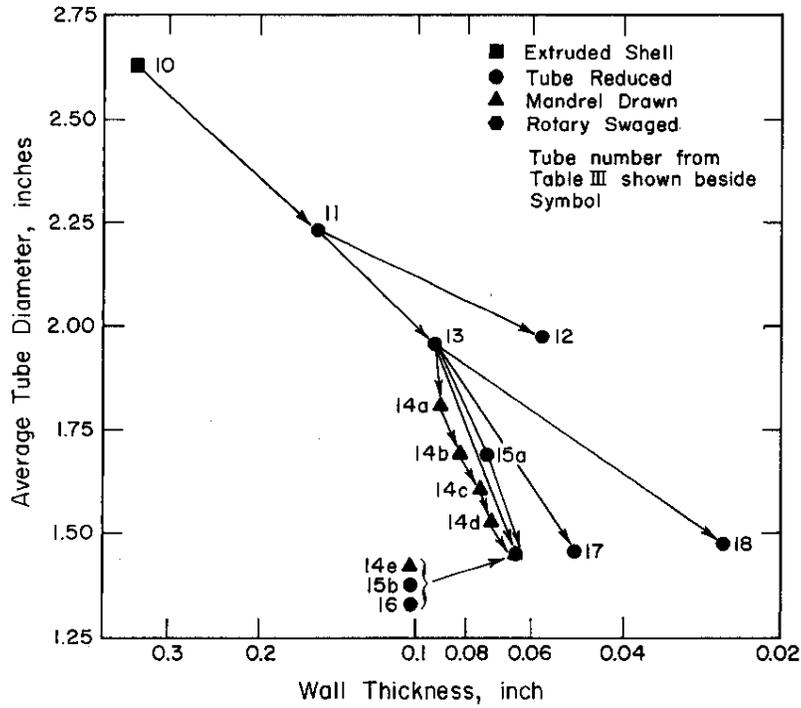


FIG. 3 FABRICATION SEQUENCES FOR SERIES-1 COMMERCIAL ZIRCALOY-2 TUBING

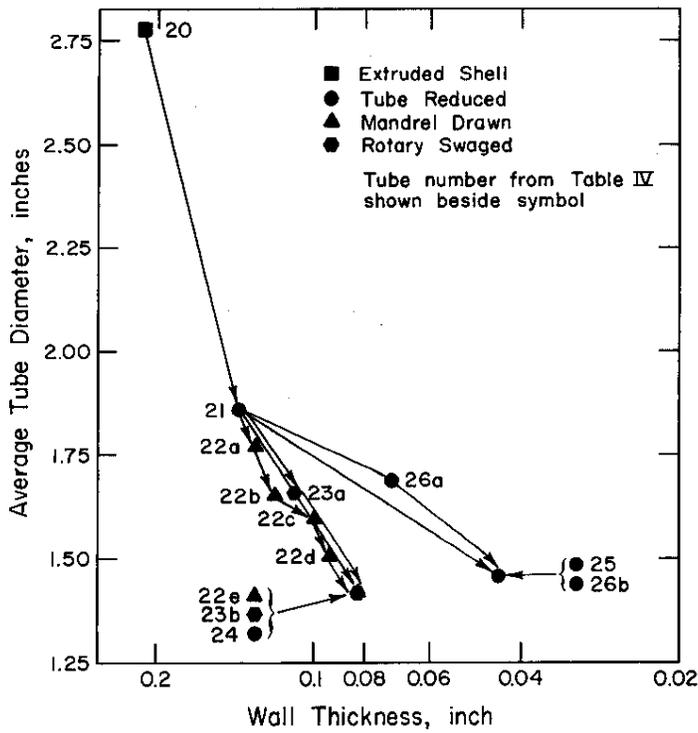


FIG. 4 FABRICATION SEQUENCES FOR SERIES-2 COMMERCIAL ZIRCALOY-2 TUBING

TABLE III
Data for Series 1 Commercial Zircaloy-2 Tubing

Tube	Process	Average Diameter \bar{D} , in.	Wall Thickness W, in.	$\Delta\bar{D}$, %	ΔW , %	DEP [$\Delta\bar{D}-\Delta W$], %	Hydride Orientation ^(a) f_{48} , %
	Billet	5.085	2.685	-	-	-	-
10	Extrude	2.628	0.352	-49	-87	+38	45
11	Tube reduce	2.221	0.154	-15.5	-56	+40	24
12	Tube Reduce from 11	1.942	0.058	-12.5	-62	+50	15
13	Tube Reduce from 11	1.905	0.092	-14.2	-40	+26	21
14a	Draw from 11	1.811	0.091	-4.9	-1.1	-4	24
14b	Draw from 14a	1.685	0.085	-7.0	-6.6	0	40
14c	Draw from 14b	1.608	0.077	-4.6	-9.4	+5	33
14d	Draw from 14c	1.525	0.072	-5.2	-6.5	+1	35
14e	Draw from 14d	1.434	0.066	-6.0	-8.3	+2	41
15a	Tube Reduce from 13	1.675	0.077	-12.1	-16.3	+4	25
15b	Tube Reduce from 15a	1.437	0.065	-14.2	-15.6	+1	36
16	Tube Reduce from 13	1.438	0.065	-24.5	-29.4	+5	35
17	Tube Reduce from 13	1.451	0.050	-23.8	-45.7	+22	29
18	Tube Reduce from 13	1.475	0.026	-22.6	-72	+49	17

(a) f_{48} = percent of hydride platelets oriented between 48° and 90° to the circumferential direction.

TABLE IV
Data for Series-2 Commercial Zircaloy-2 Tubing

Tube	Process	Average Diameter \bar{D} , in.	Wall Thickness W, in.	$\Delta\bar{D}$, %	ΔW , %	DEP [$\Delta\bar{D}-\Delta W$], %	Hydride Orientation ^(a) f_{48} , %
	Billet	5.238	2.474	-	-	-	-
20	Extrude	2.779	0.218	-47	-91	+44	47
21	Tube Reduce	1.862	0.139	-33	-36	+3	56
22a	Draw from 21	1.776	0.129	-4.6	-7.2	+3	72
22b	Draw from 22a	1.657	0.119	-6.7	-7.7	+1	69
22c	Draw from 22b	1.590	0.100	-4.0	-16	+12	59
22d	Draw from 22c	1.505	0.095	-5.3	-5.0	0	63
22e	Draw from 22d	1.419	0.083	-5.7	-12.6	+7	59
23a	Swage from 21	1.652	0.113	-11.3	-18.7	+7	64
23b	Swage from 23a	1.424	0.078	-13.8	-31	+17	64
24	Tube Reduce from 21	1.427	0.080	-23	-42	+19	61
25	Tube Reduce from 21	1.459	0.045	-22	-68	+46	40
26a	Tube Reduce from 21	1.681	0.073	-10	-47	+37	44
26b	Tube Reduce from 26a	1.458	0.045	-13	-38	+25	31

(a) f_{48} = percent of hydride platelets oriented between 48° and 90° to the circumferential direction.

The previous study of fabrication strains⁷ indicated that the $D\epsilon P$ of the last major fabrication sequence was the primary variable in determining the predominant hydride orientation, and that earlier fabrication history was of minor significance. The hydride orientations produced in Series-1 and -2 tubing are shown in Figures 5 and 6 with the $D\epsilon P$ of the last fabrication pass as the independent variable.

Figures 5 and 6 show that hydride orientations were quite different in the two series of tubes. The hydride orientations produced in Series-1 tubes were more circumferential (lower f_{48}) than expected from the baseline relationship,⁷ while the hydride orientations produced in Series-2 tubes were more radial than expected. These results were apparently related to the directional strain history of the *base tubes*, Tube 3 in Series 1 and Tube 1 in Series 2. This difference between the two series of

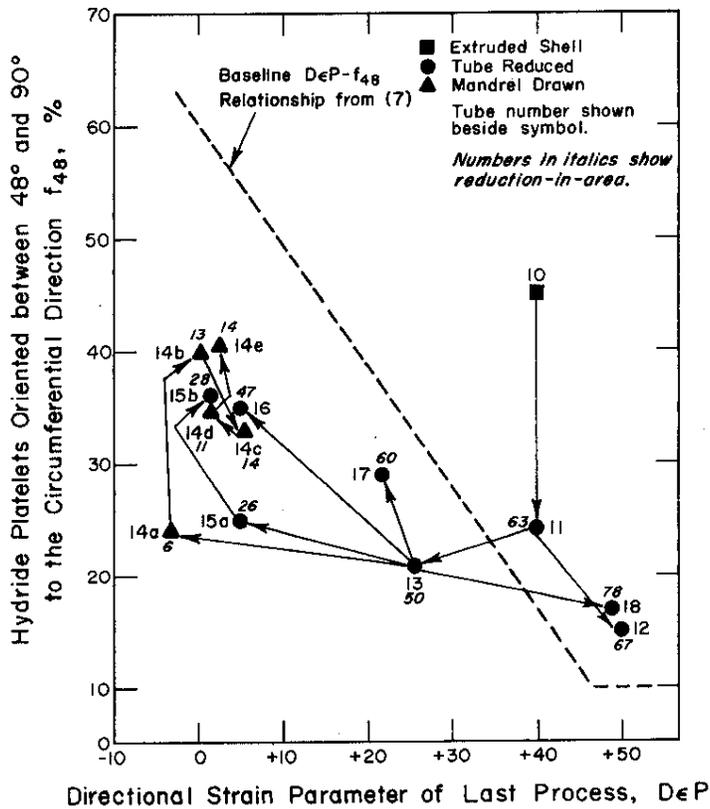


FIG. 5 HYDRIDE ORIENTATIONS IN SERIES-1 ZIRCALOY-2 TUBING (Initially "good" tube downgraded by "bad" processing steps)

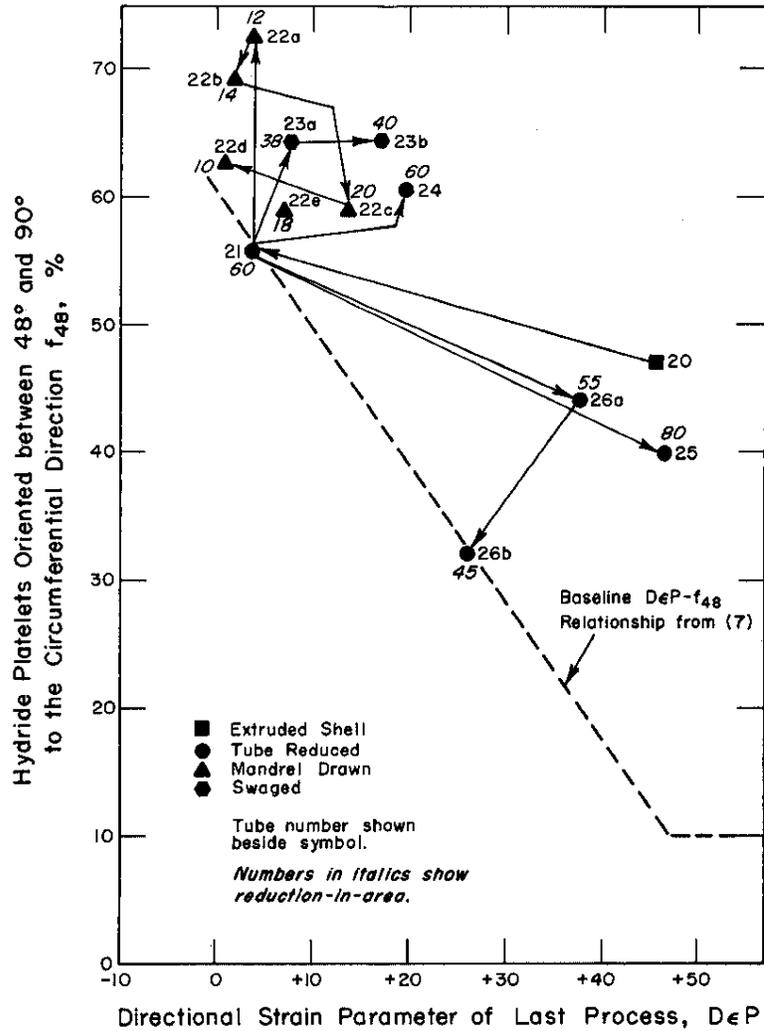


FIG. 6 HYDRIDE ORIENTATIONS IN SERIES-2 ZIRCALOY-2 TUBING (Initially "bad" tube improved by "good" processing steps)

tubes was contrary to our earlier conclusion, and showed that the strain history preceding the last fabrication step was also a major factor in determining hydride orientation.

Detailed examination of the f_{48} data for the two series of tubes added additional support to the total fabrication history concept of hydride orientation control. For example, "low DeP " processing caused f_{48} to increase in both series, although f_{48}

for the base tubes was 21 in Series 1, Tube 3* (13) and 46 in Series 2, Tube 1 (21). Also significant in this respect were the absolute values of f_{48} resulting from similar low DεP processing in the two series: the (14) mandrel-drawn tubes exhibited f_{48} values of 30-40% while the (22) mandrel-drawn tubes had f_{48} of 60-70%. Our original final fabrication DεP concept of hydride orientation control would have predicted similar values of f_{48} for Tubes 14E and 22E because of the similar processing. Similarly modified conclusions resulted from comparing Tubes 18 and 25, which exhibited hydride orientations (f_{48}) of 17 and 40%, respectively; they both had been tube reduced by almost identical amounts, 78 and 80% reduction in area, and by identical changes in average diameter and wall thickness (Tables III and IV). These data indicated, therefore, that the total fabrication history was important in determining hydride orientation.

The hydride orientation data were then reanalyzed, this time comparing hydride orientation to total, cumulative directional strain. This analysis, Figure 7, showed some scatter in the data for each series of tubing but indicated a good fit between the two series. Both series of data showed that high values of total DεP produced highly circumferential hydride orientations (low f_{48}). As seen from Table III, the only sequence of *decreasing* total DεP (tube sequence 13-14a-14b) increased the percent of radial hydrides from 21 to 40. On the other hand, higher f_{48} values were produced also by small *increases* in total DεP: tube sequences 20-21-22a, 21-23a, 13-15a-15b, and 13-16. These data showed that the sense of the DεP for each sequence of process steps was not in complete agreement with the direction of the corresponding changes in f_{48} , which indicated that a modification or refinement in the statement of DεP was necessary.

*For convenience, Series 1, Tube 3 will be designated as (13).

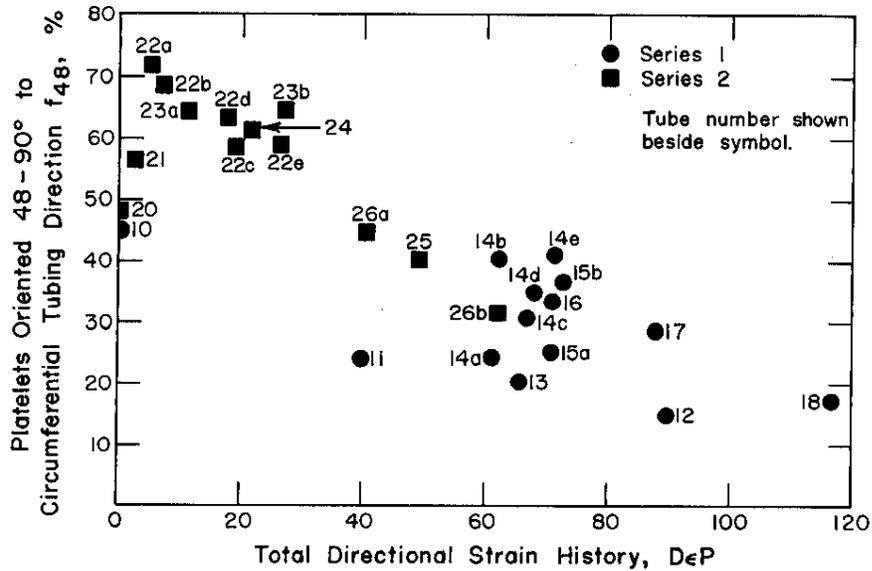


FIG. 7 HYDRIDE ORIENTATION AS RELATED TO TOTAL STRAIN HISTORY (Commercial Tubing - Transverse Plane)

RESULTS

REFINEMENT OF $D\epsilon_P$

The required refinement of the directional strain parameter was determined by analysis of the fabrication and hydride orientation data for four specific sets of tubes. The processing and dimensional changes of these tubes, listed in Table V, had been designed into the fabrication program to provide samples for one-variable analyses, i.e., the effects of various amounts of wall reduction at constant diameter reduction. The data for the fifth "set" listed in Table V is extremely pertinent to the present discussion because the "23b" process caused no substantial change in f_{48} . The data in Table V show that processes with wall changes less than twice the diametral changes produced increases in f_{48} while processes with wall changes more than twice the diametral changes produced decreases in f_{48} . Little effect of fabrication process or prior value of f_{48} could be seen. Thus, reductions in wall thickness were about half as effective in controlling hydride orientation as reductions in diameter, indicating that only a fraction of the wall-thickness reductions should be included in a new definition of the directional strain parameter.

TABLE V
Data Sets for Comparison of $\Delta\bar{D}$ and ΔW

Prior History		Last Fabrication Step						Major Variable
Tube	$f_{48}, \%$	Tube	Processes ^(a)	$\Delta\bar{D}, \%$ ^(b)	$\Delta W, \%$ ^(b)	$\Delta f_{48}, \%$	$f_{48}, \%$	
13	21	16	T.R.	-25	-29	35	+14	ΔW
13	21	17	T.R.	-24	-46	29	+8	ΔW
13	21	18	T.R.	-23	-72	17	-4	ΔW
21	56	24	T.R.	-23	-42	61	+5	ΔW
21	56	25	T.R.	-22	-68	40	-16	ΔW
21	56	24	T.R.	-23	-42	61	+5	$\Delta\bar{D}$
21	56	26a	T.R.	-10	-47	44	-12	$\Delta\bar{D}$
21	56	23a	S	-11	-19	64	+8	Process and ΔW
21	56	26a	T.R.	-10	-47	44	-12	Process and ΔW
23a	64	23b	S	-14	-31	64	0	-

(a) Fabrication processes. T.R. - tube reduction; S - swage
 (b) Dimensional data have been rounded to facilitate inspection

A quantitative definition of D_{EP} as well as the effects of zirconium grain size and hydride location was determined by multi-variable computer analyses of these data. The computer results will be described after a qualitative description of the grain size and hydride location effects.

Effect of Grain Size

Grain sizes were measured on photomicrographs of the transverse tubing plane by lineal analysis with a scribed circle to eliminate bias due to oriented, elongated grains. The apparent grain diameters calculated from lineal analysis were recorded as a Mean Planar Grain Diameter (MPGD) in Table VI, rather than being converted to a grain size number; grain size varied from 0.005 to 0.029 mm. No effect of grain size could be observed on the hydride orientation Directional Strain relationships described in Figures 5, 6, and 7, even when the definition of D_{EP} was varied in attempts to produce a grain size effect.

TABLE VI
Orientation and Location of Hydride Platelets
in Commercial Zircaloy Tubing

Tube	MPGD, ^(a) 10 ⁻³ mm	Total Hydride Platelets	Transgranular Hydride Platelets		Grain Boundary Hydride Platelets	
		f ₄₈ , %	Fraction	f ₄₈ , %	Fraction	f ₄₈ , %
10	16	45	0.68	51	0.32	33
11	9.7	24	0.76	21	0.24	33
12	8.2	15	0.72	11	0.28	23
13	11	21	0.65	20	0.35	20
14a	11	24	0.76	20	0.24	36
14b	20	40	0.65	38	0.35	44
14c	20	33	0.63	32	0.37	35
14d	19	35	0.60	34	0.40	35
14e	15	41	0.69	40	0.31	44
15a	15	25	0.70	24	0.30	27
15b	16	36	0.62	33	0.38	41
16	11	35	0.78	36	0.22	34
17	11	29	0.89	30	0.11	28
18	8.4	17	0.71	13	0.29	25
20	5.0	47	0.72	54	0.28	28
21	8.0	56	0.73	59	0.27	48
22a	29	72	0.70	73	0.30	70
22b	15	69	0.75	71	0.25	65
22c	23	59	0.63	68	0.37	44
22d	19	63	0.70	69	0.30	45
22e	21	59	0.58	63	0.42	52
23a	11	64	0.63	70	0.37	54
23b	12	64	0.72	67	0.28	56
24	6.5	61	0.69	60	0.31	54
25	7.6	40	0.70	42	0.30	36
26a	8.4	44	0.73	43	0.27	47
26b	9.7	31	0.72	31	0.28	31

(a) Mean Planar Grain Diameter

Grain size did not affect the percent of hydride platelets in the grain boundaries, e.g., in Table VI, Tubes 24, 25, 26a, 26b, 22a, 14e, and 15a all exhibited close to 70% of the platelets within the grains while the MPGD varied from 0.0064 to 0.029 mm.

Effect of Hydride Location

For the commercially fabricated series of tubes, the location of the hydride platelets, i.e., within the zirconium grains or in the grain boundaries, was determined at the time the platelet orientation was measured. Analysis of the data included calculation of frequency distributions and f₄₈ values for both transgranular and intergranular hydride platelets, and the percent of platelets in each location.

The orientation properties of the transgranular and intergranular platelets were similar in both $f_{4,8}$ values and the frequency distribution of orientations. The similarity in frequency distributions for these platelets is illustrated in Figure 1 for two tubes with widely different hydride orientation properties.

The majority, typically 70%, of hydride platelets were located within the grains (Table VI). The percent of transgranular platelets, which varied between 58 and 89%, had no observable effect on the preferred orientation, $f_{4,8}$, of the total hydride population.

STATISTICAL ANALYSIS

The data correlations presented thus far have tested the relative effect of four variables on hydride orientation: the fabrication strains, $\Delta\bar{D}$ and ΔW , were shown to be major controlling factors, while zirconium grain size and the location of the hydride platelets were shown to be minor factors. Computer analyses determined quantitatively the effect of each factor and its statistical significance on hydride orientation. The $f_{4,8}$ data was compared by multivariable, least square analyses to the paired values of:

$\Delta\bar{D}$, total change in average diameter after extrusion

ΔW , total change in wall thickness after extrusion

L, hydride location, as fraction of hydrides within zirconium grain

G, zirconium grain size, as mean planar grain diameter (mm)

The computer calculated the linear regression equation

$$f_{4,8} = C_0 + C_1 \Delta\bar{D} + C_2 \Delta W + C_3 G + C_4 L \quad (1)$$

for each series of commercial tubing, individually, and for the combined data of the two series. The data could be combined as one sample because the beginning points, $f_{4,8}$, of the two extruded

shells were similar. In addition to the regression coefficients, C_1 __ C_4 , for each of the four independent variables, the computer calculated:

- the standard error and the significance coefficient (t) of each regression coefficient,
- the multiple correlation coefficient
- the significance coefficient (F)
- the standard error of the estimate for each regression equation.⁷

The results of the statistical analysis of the two series of commercial tubing as one data sample will be described in detail since it was the most complete sample in terms of magnitude and directions of changes in hydride orientation during fabrication. These data are given in Table VII. Analyses of each series separately resulted in slightly different regression coefficients but no improvement in multiple correlation coefficients and standard errors of the estimates.

Significance of Parameters

The comparison of hydride orientation to fabrication strains alone, the first set of statistical data in Table VII, showed a very high degree of correlation, since the regression equation using only $\Delta\bar{D}$ and ΔW explained 92.7% of the variations in hydride orientation, $f_{4.8}$.

Significance of Zirconium Grain Size and Hydride Location

The addition of zirconium grain size to the multiple correlation as a third independent variable caused little change in the statistical factors (see Table VII).

A similar analysis showed that including hydride location in the multiple correlation caused essentially no change in the statistical factors.

TABLE VII

Statistical Data from Analysis of Hydride Platelet Orientations in Commercial Tubing

$$F_{4,9} = C_0 + C_1 \Delta \bar{D} + C_2 \Delta W + C_3 G + C_4 L$$

C ₀	Change in AVG Dia ΔD			Change in Wall Thickness ΔW			Zirconium Grain Size G			Location of Hydride Platelets L			Coefficient of Multiple Correlation R _{4,9}	Total Explained Variation in F _{4,9} R ² F _{4,9}	Standard Error of Estimate S _{F_{4,9}}	Significance Factor of F _{4,9} F	Probability of F P
	C ₁	t ₁	P ₁	C ₂	t ₂	P ₂	C ₃	t ₃	P ₃	C ₄	t ₄	P ₄					
41.6	-1.11	11.3	<10 ⁻³	0.529	15.8	<10 ⁻³	-	-	-	-	-	-	0.963	0.927	0.050	139	<10 ⁻³
36.6	-1.07	11.4	<10 ⁻³	0.509	15.8	<10 ⁻³	3.74	2.2	0.04	-	-	-	0.970	0.941	0.046	110	<10 ⁻³
23.7	-1.08	11.5	<10 ⁻³	0.511	15.9	<10 ⁻³	4.39	2.4	0.03	0.167	1.1	0.30	0.972	0.945	0.046	84	<10 ⁻³

NOTES:

1. 25 samples used in analysis.
2. C₁-C₄ are regression coefficients of independent variables.
3. t₁-t₄ are significance factors for regression coefficients.
4. P₁-P₄ are probabilities of obtaining "t" with 25 samples by random sampling.
5. Units for independent variables: ΔD and ΔW, percent; G, mm; L, fraction in grains.
6. Statistical terms are defined in Reference 9.

DEFINITION OF DεP-II

During the description of the hydride orientation data for the two sets of commercial tubing, page 18, wall thickness reductions were shown to control f_{48} less than reductions in average diameter did; thus, DεP needed to be modified. The DεP was modified using the coefficients from the regression equations relating hydride orientation to fabrication strains (Table VII). The relative effects of $\Delta\bar{D}$ and ΔW was obtained by normalizing the regression coefficients to a $\Delta\bar{D}$ coefficient to 1.00, i.e., by dividing, respectively, the coefficients for $\Delta\bar{D}$ and ΔW , C_2 and C_3 , by the $\Delta\bar{D}$ coefficient, C_1 . For example, the three-variable regression equation from Table VII was converted to

$$f_{48} = 36.6 - 1.07[\Delta\bar{D} - 0.477 \Delta W - 0.351 GS] \quad (2)$$

For the three equations given in Table VII, the three normalized coefficients of ΔW were 0.475, 0.477, and 0.474, showing that the *comparative* effect of the $\Delta\bar{D}$ and ΔW fabrication strains was constant and unaffected by the inclusion of additional variables. These values, which agreed well with the earlier qualitative analysis of the f_{48} data, page 18, indicated that when tube fabrication is accomplished by biaxial deformation, reductions in average diameter cause about twice the change in hydride orientation as that caused by reductions in wall thickness. Differences between predicted (regression equation) and observed hydride orientations showed no relation to fabrication process, indicating that the comparative effects of $\Delta\bar{D}$ and ΔW on hydride orientation were similar for the three fabrication processes tested: tube reduction, mandrel drawing, and rotary swaging.

These studies indicated that $\Delta\bar{D} - 0.47 \Delta W$ is the basic component describing the effect of fabrication strains on hydride orientation. Since there is other reported evidence⁴ that grain size does exert some control over hydride orientation, Equation 2 should provide the best prediction of hydride orientation using DεP-II. In those cases where the range of grain size is limited, however, the slight increase in accuracy in predicting hydride

orientation may not be worth the extra work needed to obtain the grain size data, and the two-variable equation would be satisfactory,

$$f_{4.8} = 41.6 - 1.11[\Delta\bar{D} - 0.475 \Delta W] \quad (3)$$

Tests of DEP-II

Fabrication data for two other sets of commercial tubes were available for qualitative tests of DEP-II. Tubes A and C from earlier work⁷ had both been fabricated from the same extruded base tube. Following extrusion, Tube C was tube reduced in two passes; Tube A was tube reduced two passes and then roll formed to further reduce the wall thickness with little change in average diameter. The fabrication data for each of the two tubes and DEP-II, as expressed in Equation 3, were used to back-calculate the hydride orientation of the common extruded base tube. From Tube A data, the extruded tube was calculated to have had an $f_{4.8}$ of 57%; from Tube C data, the extruded tube had a calculated $f_{4.8}$ to 60%. This is good agreement considering that only the first tube reducing pass was common to their fabrication histories.

The fabrication data for the other tubes described in Reference 7 were used as another test of DEP-II. The fabrication data for three completely different tubes were used to predict the $f_{4.8}$ that had been exhibited in each extruded tube before fabrication. These values ranged from 48 to 70%, each a very rational value for extruded tubing.

CONCLUSIONS

The results of this study showed that the natural hydride orientation in thin-walled Zircaloy tubing was controlled by its total directional strain history. Hydride location and tubing grain size played very minor roles in the natural hydride orientation and for practical purposes can be neglected.

The previously reported directional strain parameter $D\epsilon P = [\Delta\bar{D} - \Delta W]$ was refined and a new parameter, $D\epsilon P-II$, was defined as $[\Delta\bar{D} - 0.475 \Delta W]$. $D\epsilon P-II$ explained 93% of the variation in platelet orientations and therefore can readily be used to predict the influence of various fabrication steps on the natural hydride orientation in thin-walled tubing.