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THE ZIRFLEX DECLADDING OF TUBE-IN-TUBE
TYPE FUEL ELEMENTS

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INTRODUCTION

In reviewing the applicability of Zirflex process to decladding oxidized Zircaloy from tube-in-tube type, uranium fuel elements processing problems such as gas blanketing and poor diffusion in the annuli have been visualized. A pilot unit study for the definition of these potential problems is reported here.

SUMMARY

Two pilot unit Zirflex dissolutions were carried out on near prototypical tube-in-tube type elements clad in oxidized Zircaloy. The runs were made with the elements horizontal and at simulated large scale operating conditions. The results may be summarized as follows.

1. No significant difference was noted between the actual decladding achieved in these experiments and that which was predicted from prior studies(1,2) on similarly oxidized elements with somewhat different geometries.
2. There was no gas blanketing nor diffusion effects observed.
3. Initially, preferential attack was noted on areas where oxide film had been scratched or handled. However, near the end of a run a random distribution of undissolved cladding existed.
4. End plugs dissolved rapidly as had been noted in earlier Zirflex studies.
5. Ninety percent of the cladding was removed in 6.5 hours as opposed to 100 percent dissolution attained in 6 hours in earlier studies of similarly oxidized elements. The difference is attributed to an inadequate dissolvent charge resulting from a low estimate of the quantity of Zircaloy cladding. In addition, the relatively low boilup rate used contributed to slowing the reaction due to ammonia buildup in the dissolvent.

EQUIPMENT AND MATERIALS

A Batch dissolver (A-5) formerly used for head end studies on conventional aluminum clad slugs was reactivated for the tests. It was equipped with a heating coil and a down draft condenser. A meter was attached to the condensate removal line to measure flow to a calibrated receiver tank. A head tank was connected to the dissolver through a rotameter for water replacement of removed condensate.

Chemical reagents used in the tests were technical grade ammonium fluoride and ammonium nitrate. The fuel elements were reject, tube-in-tube type, Zircaloy clad uranium obtained from FPD. They had been oxidized in an autoclave at 1500 psia and 400°C for 36 hours. The physical dimensions of the elements are given in Table I.

TABLE I
FUEL ELEMENT DIMENSIONS

	<u>OD</u>	<u>ID</u>	<u>Length</u>	<u>Clad</u>	<u>End Cap</u>
Outer tube	1.825"	1.460"	14.0"	0.022"	0.20"
Innter	1.049"	0.498"	14.0"	0.022"	0.20"

DISSOLUTION

Two pilot unit runs were made, each involving six tube-in-tube type fuel elements placed horizontally in dissolvent of initial concentration 5.5 M NH_4F - 0.5 M NH_4NO_3 . The fluoride/zirconium mole charge ratio was established at 7 in the first run and at 7.7 in the second, the latter incorporating a 10 percent increase of reagent to cope with excess cladding. As it happened, the cladding weight was underestimated so that the mole charge ratios may have been as low as 6.0 and 6.7 respectively.

The operating procedure for the runs was based on the probable method of operation in a large scale dissolver. The elements were charged to cold dissolvent which was then heated to boiling by a steam coil. When boiling started the steam pressure to the coil was adjusted to provide a constant boil-up rate. Condensate was removed to a separate receiver and replaced by continuous water addition. There was no air sparge so that dissolvent circulation was induced only by the boiling.

The run conditions and estimated dissolution are shown in Table 2. It should be noted that the first run was stopped to observe initial attack patterns while the second run was taken essentially to completion.

TABLE 2
RUN CONDITIONS

<u>Run</u>	<u>Time</u> <u>Hr</u>	<u>NH_4F</u> <u>M</u>	<u>NH_4NO_3</u> <u>M</u>	<u>F/Zr</u> <u>Mole Ratio</u>	<u>Boilup</u> <u>gm mols</u> <u>min ft²</u>	<u>Percent Dissolved</u> <u>Clad</u>	<u>End Plugs</u>
1	2.8	5.5	0.5	6-7	0.61	60	100
2	6.5	5.5	0.5	6.7-7.7	0.82	90	100

For the purpose of this study qualitative results are of more importance than quantitative measure of cladding removal. These results are shown in photographs of the cladding remaining after dissolution presented in Figures 1 through 5.

The pattern of cladding attack for Run 1 (2.8 hr) is shown in Figures 1 and 2. As would be predicted from earlier studies⁽¹⁾, outside cladding of both the inner and outer tube (areas subject to maximum handling abrasion) experienced good chemical attack. Similarly, good attack occurred on the inner cladding of

the outer tube, particularly along the strips abraded by diametrical supports during element assembly. The inside of the inner tube shows the random attack expected on unabraded Zirconium oxide surfaces. No gas blanketing or diffusion effects are apparent.

The results of Run 2 are shown in Figures 3, 4, and 5. The increase in dissolution time to 6.5 hours resulted in essentially complete (90 percent) Zircaloy removal from all four element surfaces. The random distribution of the residual cladding again indicated little or no gas blanketing or diffusion phenomena. The discrepancies in initial attack distribution on the various surfaces observed in Run 1 largely disappeared with the longer dissolution period.

REFERENCE

1. Smith, P. W., The Zirflex Process Terminal Development Report, HW-65979, August, 1960.
2. Smith, P. W., Zirflex Dissolution of the Annular Cladding of Simulated Power Fuel Elements, HW-63043, December, 1959.

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FIGURE 1

Run 1 Outer Surface of Inner and Outer Tubes

FIGURE 2

Run 1 Inner Surface of Inner and Outer Tubes

FIGURE 3

Run 2 Outer Surface of Inner and Outer Tubes

FIGURE 4

Run 2 Inner Surface of Outer Tubes

FIGURE 5

Run 2 Inner Surface of Inner Tubes