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by

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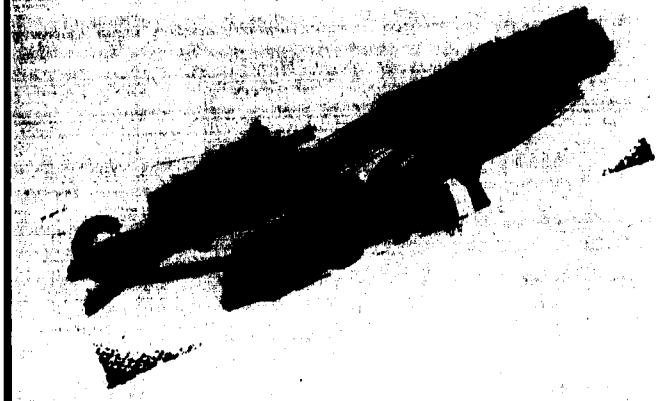
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TUNGSTEN CLADDING OF REACTOR FUELS*

by

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and R. Holzl †

This paper will be presented at the Symposium on Applications of Refractory Metals, to be held December 9 and 10, 1963, in Los Angeles, California, and will be published in the Proceedings of the Symposium.

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November 6, 1963

TUNGSTEN CLADDING OF REACTOR FUELS

INTRODUCTION

The need for cladding of reactor fuels arises because bare fuels operated at temperatures of 1800°C in reactor environments usually are not strong, often swell, suffer weight losses from vaporization, and permit a relatively large amount of fission-product release.

The fuels under consideration at General Atomic as candidate nuclear fuels to be operated at temperatures up to 1800°C are UC, UC-ZrC, W-UC, and W-UO_2 cermets. Studies at General Atomic and elsewhere of the compatibility of refractory metals and these fuels have indicated that of four metals studied (W, Ta, Mo, Nb) pure tungsten is, to date, the material most suitable for use as a direct cladding. Tungsten has considerable strength at 1800°C [1], and like other pure metals does not exhibit significant dimensional changes due to the reactor environment [2], and according to reference (3) its physical properties are not expected to be significantly changed at these temperatures. Tungsten can add significant strength to the fuels and hence enhance their dimensional stability, and can prevent the fuel element from losing weight as a result of vaporization. Further, fission-product release is impeded by the cladding.

Tungsten is a fairly difficult material to fabricate by conventional techniques. Vapor deposition was one logical choice as a method for cladding test specimens. Studies on ductile-to-brittle transition temperatures have indicated the transition temperature to be a function of grain size. Tungsten can be vapor-deposited in fine grains and, because of its resistance to grain growth, will retain its ductility and low transition temperature even when heated to temperatures higher than the recrystallization temperatures of sintered or cast tungsten.

In order to develop improved tungsten cladding of reactor fuels, General Atomic undertook a program in close cooperation with San Fernando Laboratories. Most of the vapor deposition part of the fabrication work was carried out at San Fernando Laboratories, and the research and development data analysis and fuel fabrication were done at General Atomic.

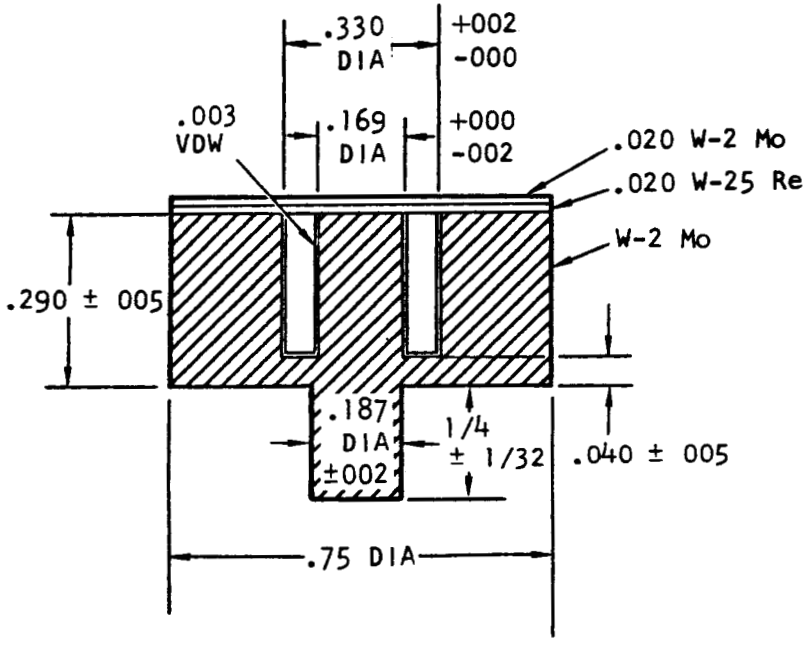
DESIGN OF FUEL-SPECIMEN CLADDING

Two different methods of tungsten cladding were employed in the studies at General Atomic. One method utilized vapor deposition, the other a machined W-2Mo alloy cup (inside surface clad with vapor-deposited tungsten) with a diffusion-bonded cap. The designs of the parts for each are shown in Figures 1 and 2. It will be noted that in both designs there is a void at the top. The void was incorporated for two reasons: (1) The void allows room for the expansion of escaped fission gases, and less stress is put on the cladding. (2) The design gives each specimen the capability of being used to measure the volume of fission gas release. Figure 3 shows a cross section of one finished vapor-deposited-tungsten-clad specimen and a top and a side view of another vapor deposited specimen. The specimen has a tungsten pin brazed in at one end. A hole was drilled in the top, as the specimen had to be out-gassed internally to remove the residue from vapor deposition; the pin seals the hole. The W-2Mo clad specimens were out-gassed internally prior to diffusion-bonding the cap to the specimen in vacuum.

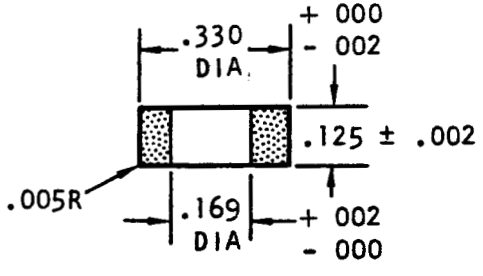
As a preliminary investigation into the feasibility of the method, several carbides were coated; the parts were out-gassed at $\sim 1800^{\circ}C$ and were sectioned and found to be intact, with no evidence of significant damage as a result of the vapor-deposition coating process. It was then assumed that the ordinary vapor-deposition techniques were feasibile for coating the carbides to be used in testing of tungsten-clad emitters.

A slight change in design of the preforms used to enclose the void was introduced to make the cladding process more economical. The first design is shown at the top of Figure 4, and the newer design, in which the tungsten preform was made in two parts and extended the length of the specimen, is shown at the bottom.

When actual cladding of the carbides to be used in the test specimens was begun, a series of unanticipated difficulties in the vapor deposition process were encountered. Every carbide reacted with the atmosphere during deposition--the degree varying from slight attack to such severe attack that the carbide turned to powder. After a number of specimens had been ruined in these attempts, it became apparent that the process would have to be changed.



CARBIDE FUEL



DETAIL B
FINISHED W-2 Mo
CLAD SPECIMEN

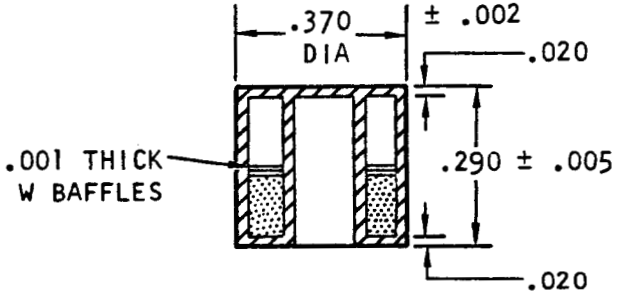
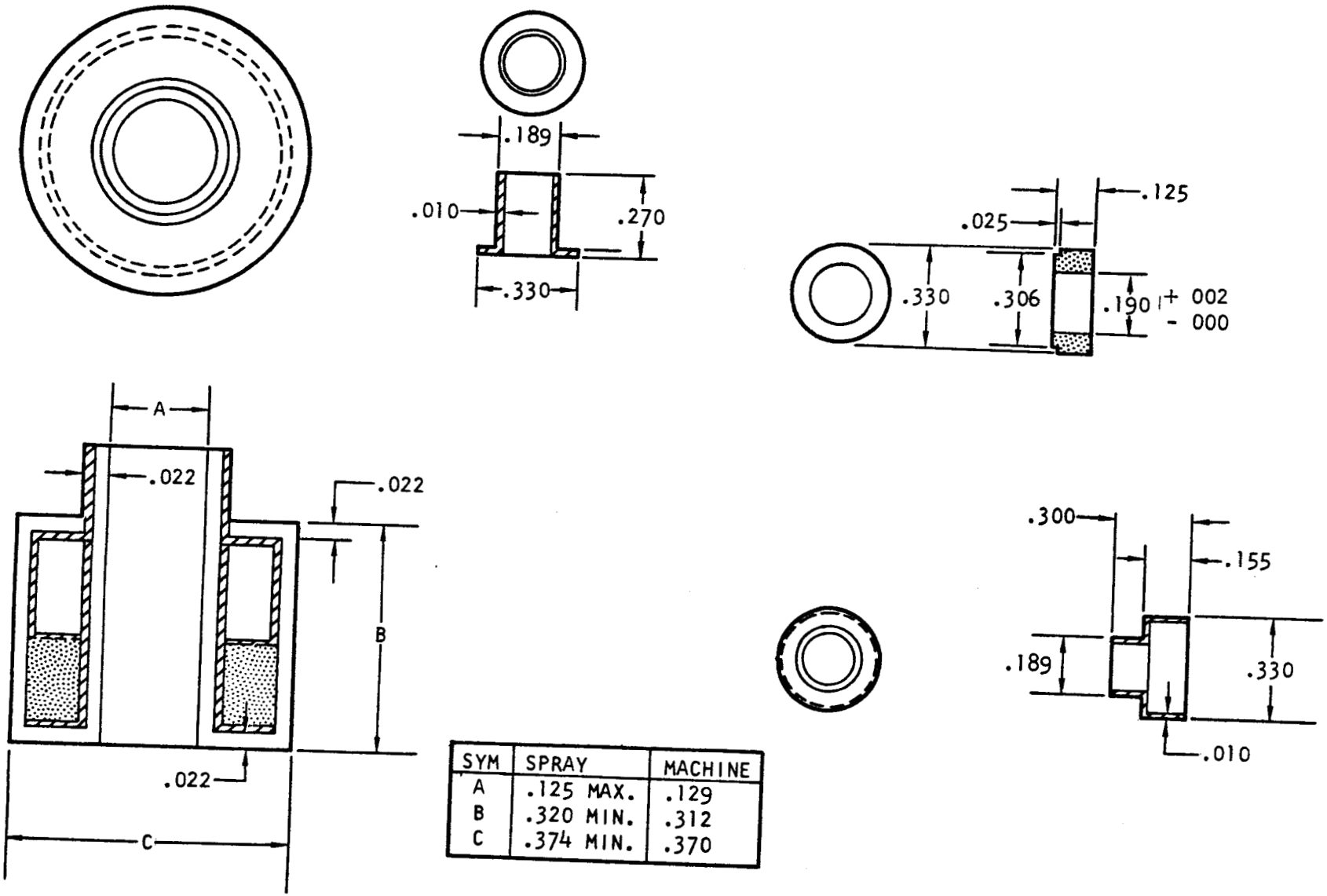
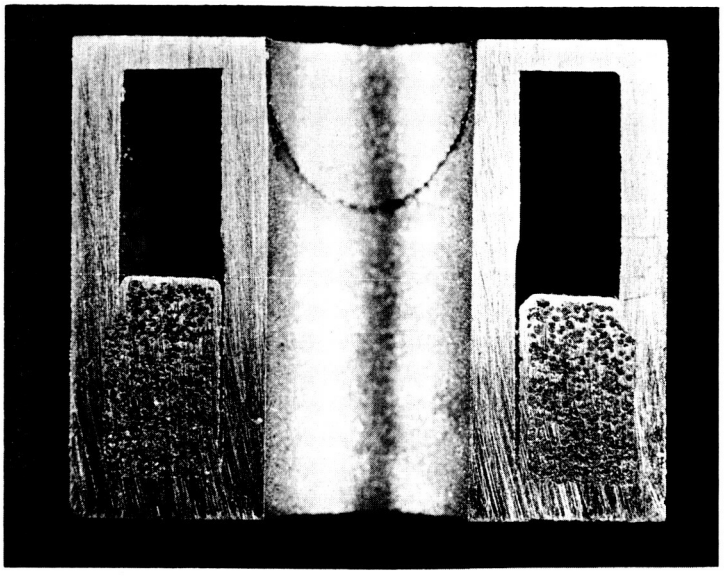


Fig. 1--W-2Mo Clad emitter specimen

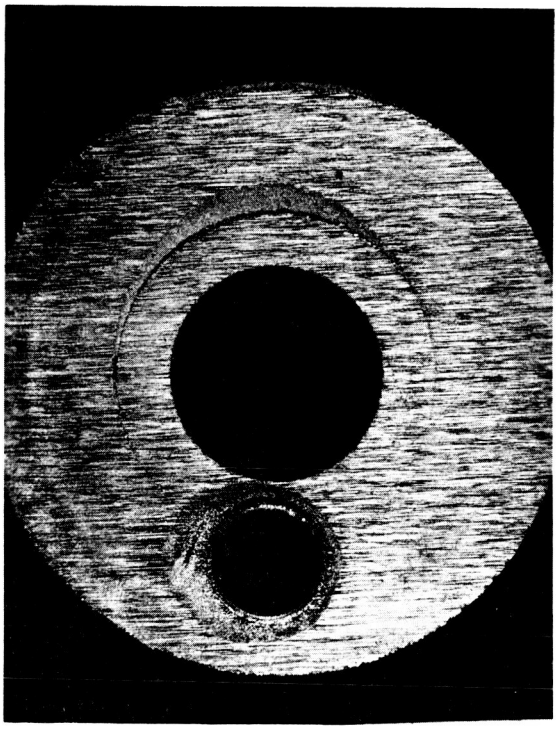


SYM	SPRAY	MACHINE
A	.125 MAX.	.129
B	.320 MIN.	.312
C	.374 MIN.	.370

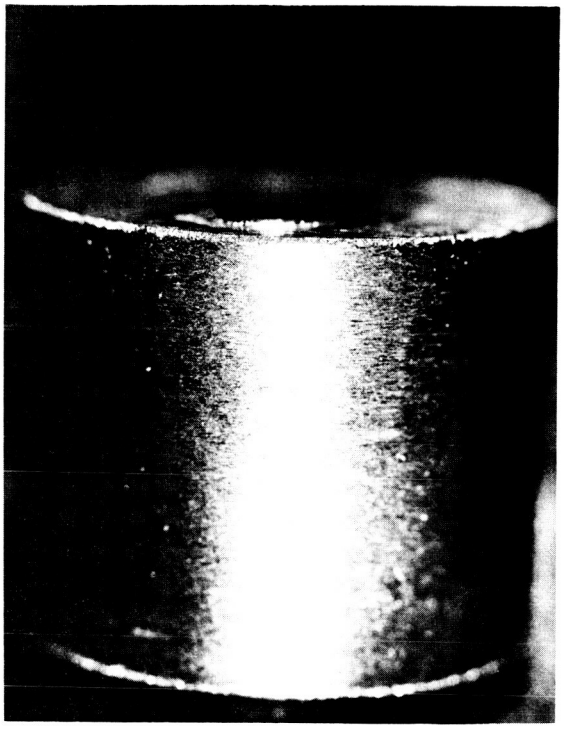
Fig. 2--Vapor-deposited W clad emitter



W-UO_2 cermet clad with vapor-deposited tungsten



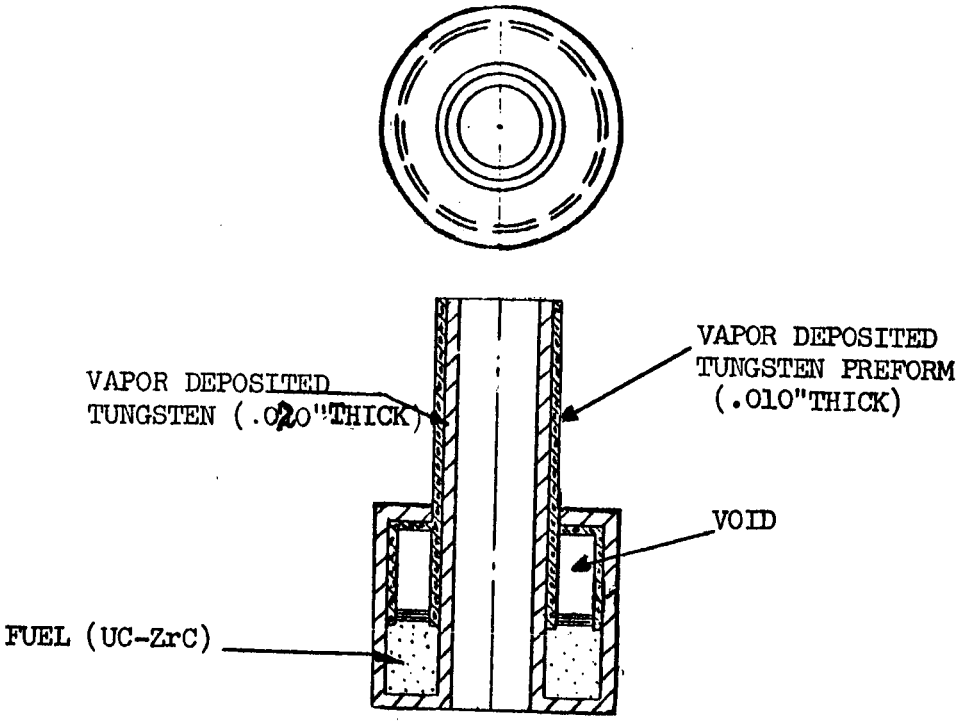
Top view



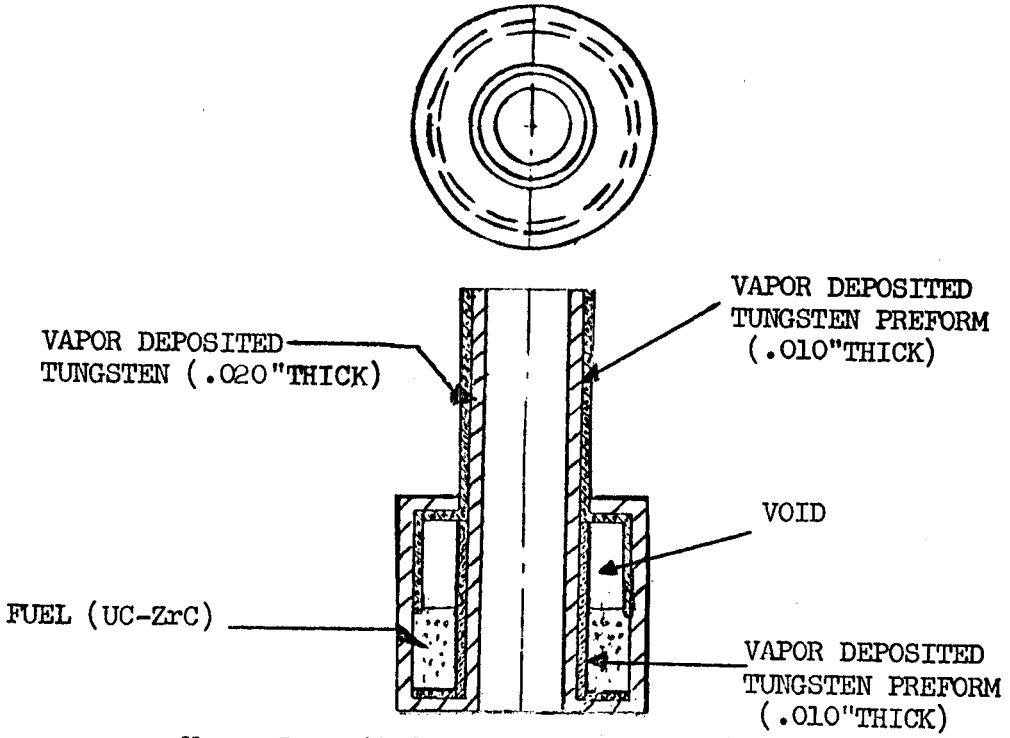
Side view

90 mol-% UC–10 mol-% ZrC (depleted) clad with vapor-deposited tungsten

Fig. 3



Vapor Deposited Tungsten Emitter (4 x 1 = scale)
(old design)



Vapor Deposited Tungsten Emitter (4 x 1 = scale)
(new design)

Fig. 4

It was also apparent that the successful coating of the carbides during the preliminary investigation was due partly to the design of the specimen (which prevented trapping of corrosive gases inside the tungsten preform) and perhaps because the plating atmosphere was cleaner than is usual during ordinary tungsten-vapor deposition. A clean atmosphere is one which contains the practical minimum of moisture, oxygen, or other contaminants.

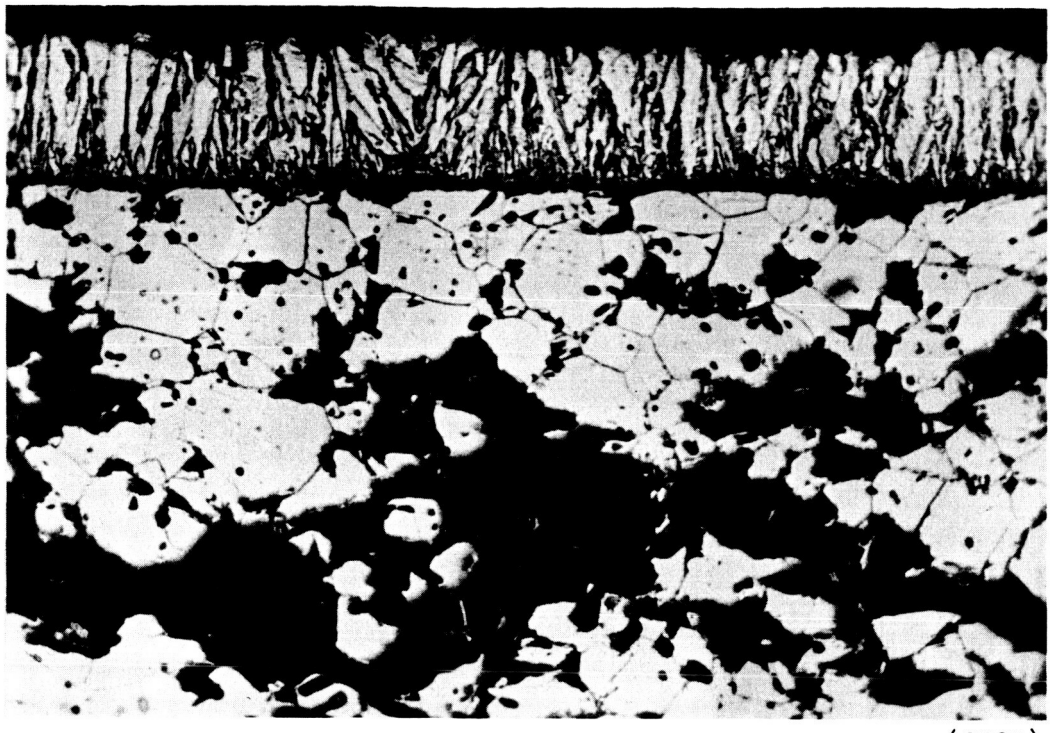
DEVELOPMENT OF SATISFACTORY CLADDING PROCEDURE

A program to develop a more suitable method for coating the carbides was started. Since UC-ZrC specimens are attacked rapidly in moist atmospheres and the attack is accelerated by raising the temperature, the first step was to minimize contamination from impurities in gases used in the plating process H_2, He, and WF_6, and to eliminate leaks in the plating system equipment.

Even after impurities were eliminated from the system it was determined that the carbides are attacked by reaction products formed during the plating operation. To prevent this type of attack on the carbides during final encapsulation, it was necessary to develop techniques for sealing the carbide specimens with a tungsten coating applied under conditions which eliminated or minimized such attack. The specimen could then be treated much like an ordinary piece of tungsten.

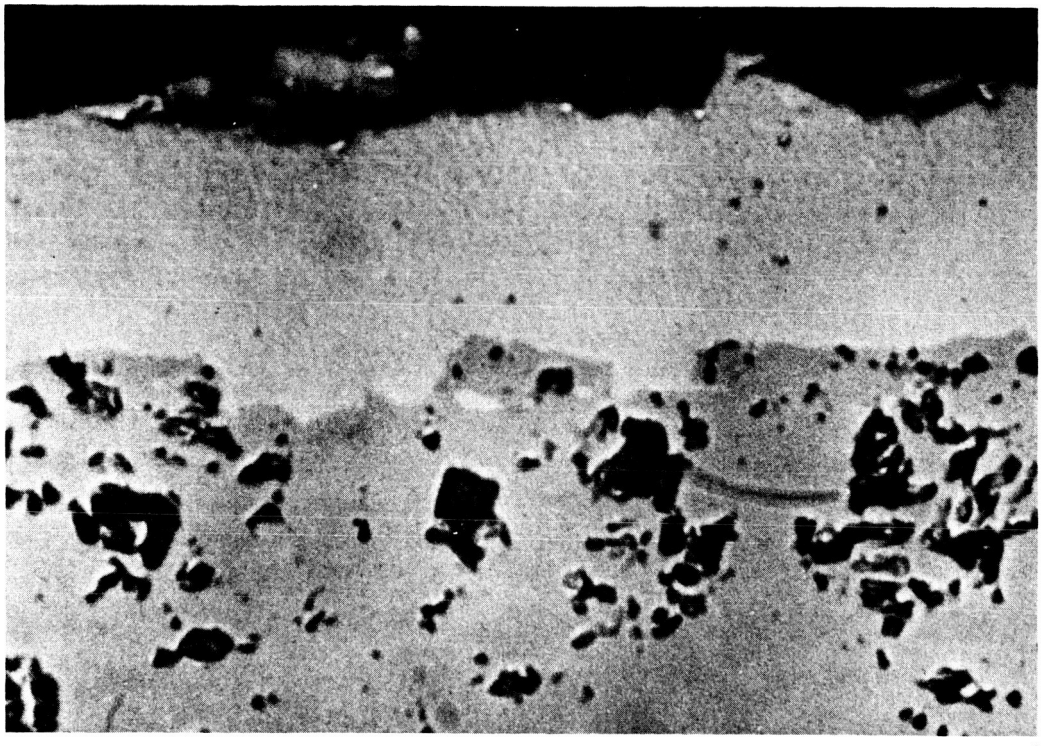
Several dozen specimens were coated under various plating conditions and evaluated by metallographic examination of the tungsten-carbide interfaces. A photomicrograph showing attack is given at the top of Figure 5; the other photomicrograph (in Figure 5) shows no significant attack. These results indicated that a vapor-deposited tungsten coating could in fact be applied with no significant attack on the carbide. When carbides with protective coatings 1-1/2 to 5 mils thick were heated in HF for one-half hour at $\sim 700^{\circ}C$, it was found that a 3-mil coating gave adequate protection provided that the coating had no holes. Figure 6 shows the setup for applying the seal coating; Figure 7 shows the arrangement for final encapsulation.

Thirty-five specimens were successfully coated by these techniques. The results showed that tungsten-vapor deposition is a fabrication method amenable to cladding of complex shapes; tungsten can be vapor-deposited



(250X)

Vapor deposited tungsten on 90UC–10ZrC specimen.
Note attack of carbide with reaction products.



(400X)

Vapor deposited tungsten on 30UC–70ZrC specimen.
Note no attack of carbide from reaction products
of vapor deposition.

Fig. 5

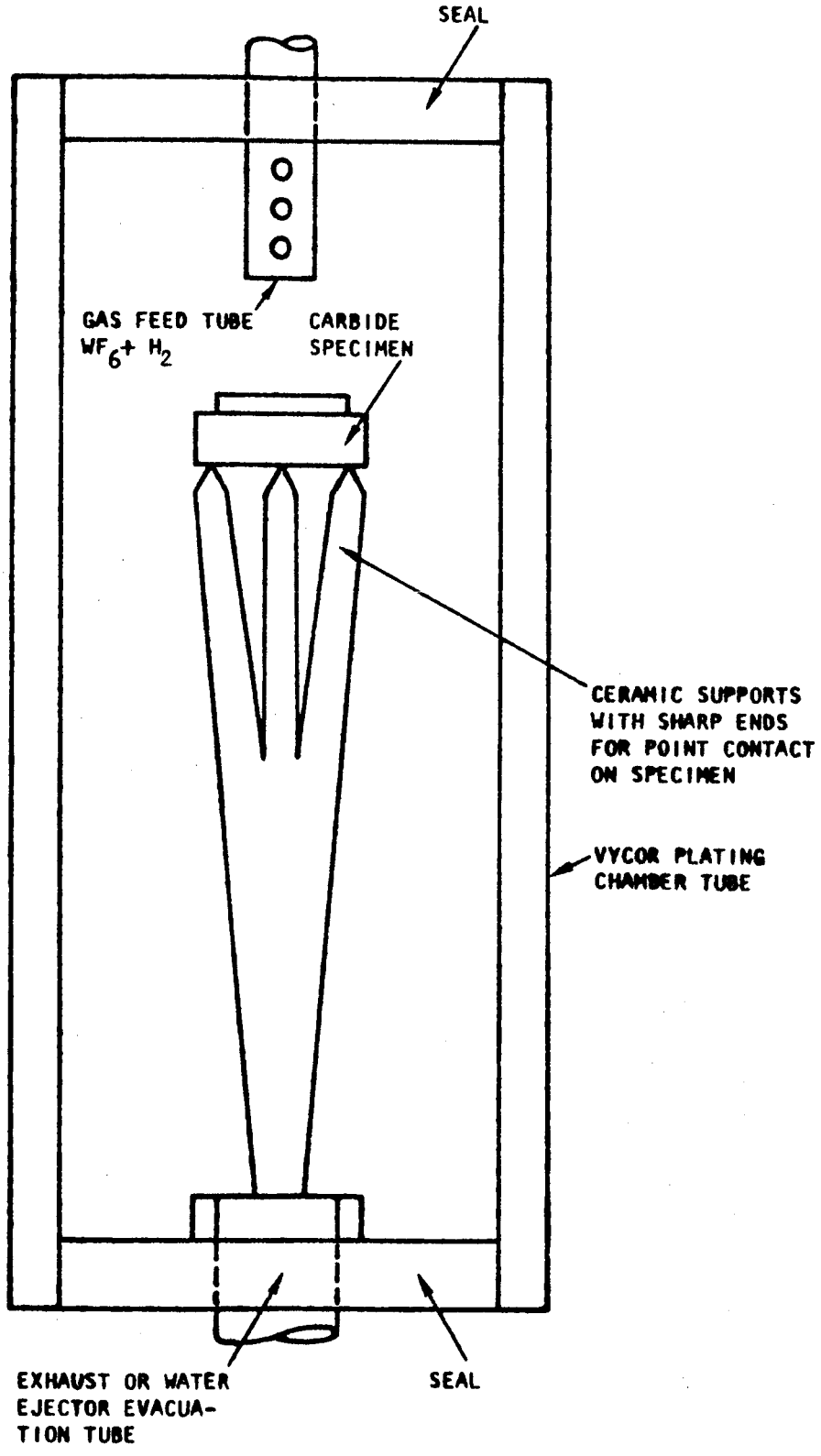


Fig. 6--Arrangement for seal coating

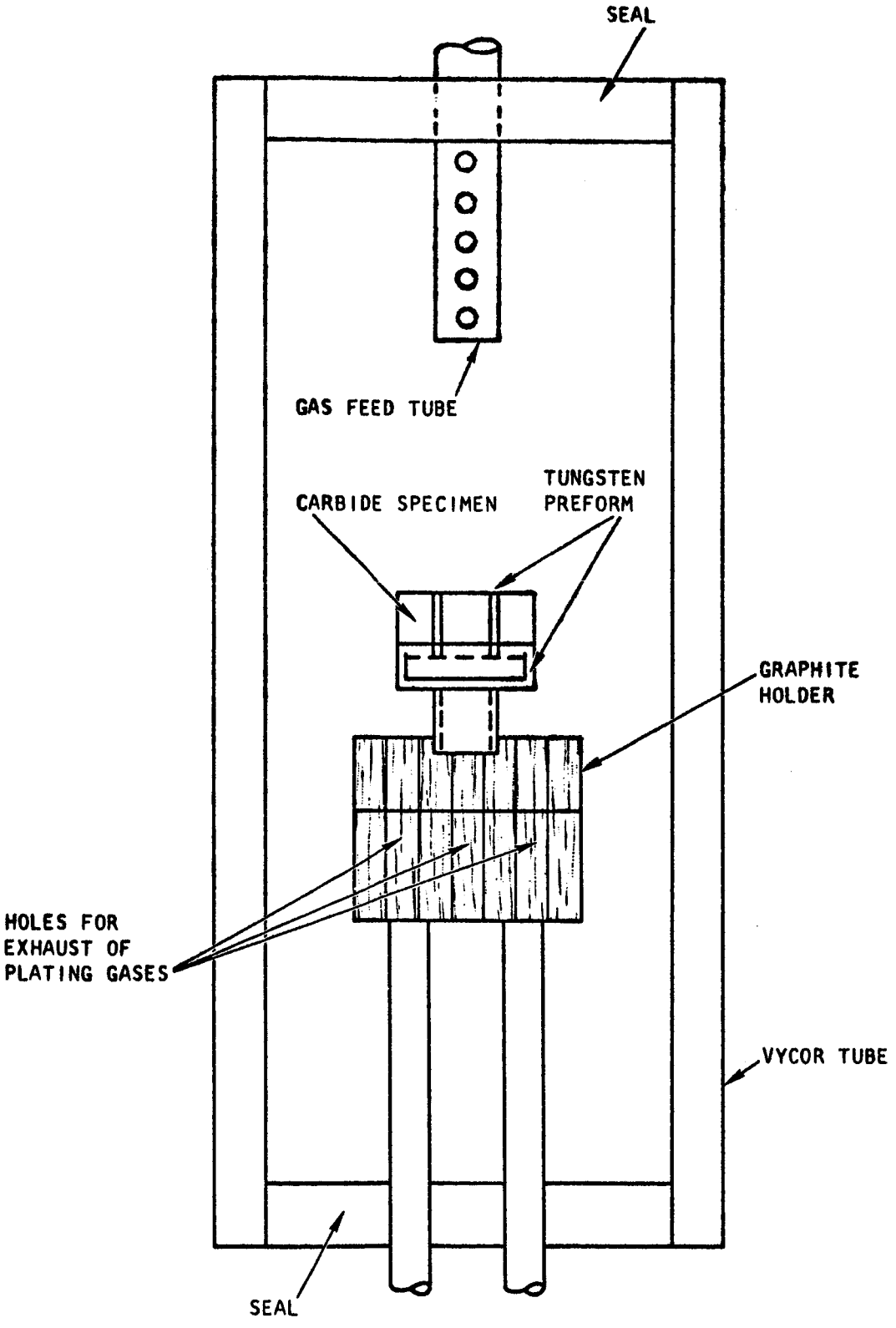


Fig. 7--Arrangement for final encapsulation

over voids such as are shown in Figure 4, and applied to extremely reactive materials such as UC. All of the specimens coated withstood machining, handling, and out-gassing without being damaged. All of the thirty-five specimens remained in tact and suitable for irradiation testing. Metallographic evaluation of several of the specimens after fabrication and out-gassing showed there was little, if any, evidence of reaction or attack on the carbide in all of the specimens examined.

References:

(1) NASA TND 1094
(2) Radiation Damage in Solids, Bellington, D. S. and Crawford, J. H. Jr., Princeton University Press, 1961
(3) Deformation, Fracture, and Radiation Damage in Body Centered Cubic Transition Metals, Johnson, A. A. Jnl. Less Common Metals, Vol. 2, pgs. 194-200 (1960)