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**CHEMICAL VAPOR DEPOSITED REFRACTORY METAL CAPSULES
FOR RADIOISOTOPE SOURCES**

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ABSTRACT

Tungsten capsules have been deposited by hydrogen reduction of WF_6 and creep tested to demonstrate their utility as containers for isotope heat sources. The capsules were subjected to stepwise pressure increases at $1650^\circ C$ (simulating helium generation of some isotopes) until failure occurred. The tangential stress at rupture was about 3300 psi and rupture occurred after a circumferential strain of 19%. These results showed that the rupture life can be predicted on the basis of the stress-temperature history.

The chemical vapor deposition (CVD) technique used to produce this pressure vessel appears to be sound, and the internal pressure test shows the deposits to be strong and ductile at the elevated temperatures.

INTRODUCTION

Tungsten is a prime candidate for encapsulating isotope heat sources in power-generating devices because the metal has high strength and thermal conductivity at high temperatures. These characteristics are needed for absolute containment of such isotope fuels as Cm_2O_3 and PuO_2 because the helium emitted by these materials through alpha decay results in high pressures. Since tungsten is difficult to fabricate by conventional methods, we have used CVD to fabricate capsules. The objectives of this study were (1) to demonstrate the feasibility of preparing tungsten capsules by CVD and (2) to evaluate CVD tungsten as a potential encapsulation material for radioisotopes. Two tungsten capsules were deposited and evaluated. Evaluation involved stress rupture of the capsules by internal pressurization at $1650^\circ C$. Since the high-temperature mechanical properties of CVD tungsten have previously been studied mostly in uniaxial tension, this study provided an opportunity to determine whether uniaxial data could be used to predict the performance of capsules.

EXPERIMENTAL

CAPSULE PREPARATION

The capsules were deposited by the hydrogen reduction of WF_6 using the deposition arrangement shown in Fig. 1. The hollow molybdenum mandrels were centered in a water-cooled silica glass tube and heated by means of an induction coil. The deposition conditions were $600^\circ C$, 3 to 4 torr, and 500 and $50\text{ cm}^3/\text{min}$ of hydrogen and WF_6 , respectively. A uniform tungsten deposit formed on the mandrels at the rate of about 0.003 in./hr. The as-deposited capsules were about 2.4-in.-long $\times$ 0.8-in.-diam $\times$ 0.04-in.-thick wall with ellipsoidal ends. A small tube (about $3/16$ in. OD) extending about 2 in. from one end was integrally deposited with the capsule. The cylindrical capsule walls were electropolished to a thickness of 0.028 in., and the molybdenum mandrels were removed by chemical dissolution. Then the dimensions of the

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Fig. 1. Apparatus for Chemical Vapor Deposition of Tungsten Capsules.

capsules were about 2.288-in.-long $\times$ 0.735-in.-ID $\times$ 0.028-in. wall. Finally, a 16-in.-long CVD tungsten tube was welded to the integrally deposited tube, providing an overall length of capsule and tube of about 20 in. Figure 2 shows the capsules in the as-deposited and the electropolished conditions. A posttest chemical analysis of one of the capsules revealed < 20 ppm C, 2 ppm H, < 1 ppm N, 28 ppm O, and 15 ppm F.

CAPSULE TESTING

Each capsule in turn was placed in a high-temperature vacuum furnace, and the tube was connected to a helium supply. The stress rupture test involved heating the capsule to 1650°C and stepwise increases in pressure of 2.5 psig/day starting with an initial pressure of 25 psig. Pressurization was continued until the capsule ruptured. There was no method for determining dimensional changes during the test; therefore, the stress during test was calculated from pretest dimensions and the strain was determined from posttest dimensions.

Fig. 2. Tungsten Test Capsules.

RESULTS

Figure 3 shows the stress history for both capsules superimposed on a plot of the uniaxial tension data for CVD tungsten obtained by McCoy and Stiegler.¹ The hatched band represents the scatter band for the uniaxial tension data, and the jagged line is the capsule data. The stress axis in Fig. 3 represents both the stress in uniaxial tension, and, in the case of the capsules, the tangential stress. The time axis represents both the duration of the uniaxial tests, and, in the case of the capsules, the time period spent at each stress level. The duration of the capsule tests, therefore, is not read directly from the time axis, but is obtained by summing each increment of time at each stress level.

The tangential, or circumferential, stress on the capsules was calculated from the expression $S_t = pD/2t$, where p is the helium pressure, D is the inner diameter, and t is the wall thickness of the capsule. Actually, D and t changed slightly during the test, but since these quantities were not continuously measured, the stress values in Fig. 3 were based on the original dimensions.

The life-fraction rule² was used in plotting the capsule data in Fig. 3. The rule states that the total lifetime of a specimen is approximated by the sum of the fractional parts of the lifetime spent at each stress level. Each horizontal portion of the capsule data plot represents the time at a given stress, and each slanted portion represents an instantaneous change in stress.

The first horizontal portion represents the time at the first (lowest) stress. At that stress the capsules had a long life expectancy based upon uniaxial data. When the stress was raised the life expectancy decreased, but, according to the rule, the fraction of the lifetime remaining at the higher stress was the same as before. The starting point for the new stress period was located at a point corresponding to the fraction of the lifetime remaining. This point can be graphically located on a line which is parallel to the uniaxial data band and intersects the end of the lower stress period. The entire plot for the capsules was prepared in this manner.

The incremental increases in stress eventually caused the capsules to fail by stress rupture. Since the two capsules were tested in an identical manner, the stress curves entered the scatter band for uniaxial data at the same time

Fig. 3. Stress History at 1650°C of Tungsten Capsules.

(73 days). A further increase in stress then caused the capsules to fail within the scatter band. The results of these tests, summarized in Table 1, show a high reproducibility in the two capsule tests.

Table 1. Summary of Results of Capsule Tests

Capsule	Rupture			
	Life (days)	Pressure (psi)	Stress (psi)	Strain (%)
1	88	257.2	3371	19.0
2	85	249.7	3273	19.3

Figure 4 shows deformation in the wall of one of the ruptured capsules and the large crack which apparently formed at the time of rupture. The crack is oriented normal to the maximum stress, which was tangential. Other small cracks can be observed nearby; presumably these cracks had not penetrated the wall at the time of failure.

A typical microstructure in the region of failure is shown in Fig. 5. Small cracks can be observed extending inward from the outer surface of the capsule. Numerous voids can also be observed in the process of linking together along grain boundaries. The nucleation and growth of grain boundary bubbles have been discussed by McCoy and Stiegler¹ and by Farrell et al.³

Fig. 4. Tungsten Capsule after Stress Rupture Test at 1650°C.

Fig. 5. Microstructure in the Region of Failure. 100x.

The voids participated in the failure of the material in two ways. First, the voids reduced the amount of material, or cross-sectional area, upon which the rupture force acted. This implies that the actual rupture stresses for the two capsules were somewhat higher than are shown in Table 1, since those values were based upon the original thickness of a fully dense wall. Second, the voids evidently contributed to the rather large tangential strain values observed, since the maximum decrease in wall thickness (around the point of rupture) was only about 0.002 in., or 7% of the original 0.028-in.-thick wall.

In the absence of voids the tangential and radial strains should be approximately equal.

CONCLUSIONS

The results of this study allow the following conclusions to be made: (1) the feasibility of preparing tungsten capsules has been demonstrated; (2) the stress rupture behavior of cylindrical capsules as used in this study can be predicted from uniaxial tension data; (3) the creep rupture properties of the two capsules tested were very similar; and (4) the large rupture stress and strain exhibited by CVD tungsten capsules would be useful for containing helium-generating isotope fuels and other applications requiring high strength and ductility at elevated temperatures.

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