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The Mechanical Properties of
Vapor-Deposited Thin Metallic Films -
A Status Report

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MASTER

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INTRODUCTION

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The mechanical properties of vapor-deposited thin metallic films are being studied in conjunction with the target fabrication group associated with the laser-fusion energy program. The purpose of the work is to gain an understanding as to which metals are structurally best suited to contain a glass microsphere filled with deuterium-tritium (D-T) gas at large internal pressures.

Initially, laser targets consisted of a thin glass microsphere containing the D-T gas at large internal pressures. The inside diameter of the sphere was about 100 μm and the thickness of glass was about 1-20 μm .⁽¹⁾ The target is placed in a vacuum chamber and subjected to high energy laser beams for a short period of time. The glass sphere vaporizes under the extreme energy of the laser transmitting a compressive shock wave to the D-T gas which, hopefully, reaches temperatures and densities needed for a fusion reaction to occur. Over the years as lasers have become higher in energy more efficient targets were needed. Figure 1 shows examples of two direct drive targets. These are a composite design consisting of the D-T gas contained by the thin glass sphere. Using vapor-deposition techniques a dense metal such as gold, tungsten or an alloy is coated onto the glass sphere to thicknesses anywhere from 10 to 50 μm . An ablative material is then deposited onto the sphere. The ablative material is used to provide a more uniform compressive shock wave to the D-T gas. A dense metal is needed primarily to prevent preheating of the gas by energetic electrons given off by the ablative material during vaporization. Preheating of the gas might

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result in a nonuniform thermonuclear burn, as well as, making the gas more difficult to compress. Also, compressing the gas to attain the densities needed for a fusion reaction to occur is facilitated by the use of a high density metal. Glass, also, has a greater probability of contaminating the gas by impurities than does metal. For these reasons the glass is becoming less important and eventually will be replaced by metal of a high atomic number.

Prior to the actual test the filled targets are stored at ambient temperatures. Under these conditions the confined D-T gas reaches an internal pressure between 100 and 200 atms. As the glass spheres are made thinner the metal becomes a critical structural component in containing the high pressure gas. As such, it becomes essential to characterize and control the mechanical properties of the metal. It would be advantageous to design a mechanical test which as closely as possible approximates the actual in-service condition. Since it is not practical to test the metal while on the sphere a reasonable facsimile is needed.

Theory

Basically the filled microsphere approximates a small pressure vessel. The stress-state in the metal is determined by D/t ratio, where D = internal diameter of the sphere and t = thickness of the metal coating. Timoshenko,⁽²⁾ has shown that for $D/t \geq 20$ the thin-walled, or membrane, theory applies in determining the stress-state in the metal,

and for $D/t < 20$ the thick-walled theory is needed. That is, a thin-walled sphere under internal pressure, experiences a tangential or hoop-stress σ_θ , a meridional stress σ_ϕ , and a radial stress σ_r . The tangential stress and meridional stress are equal due to the symmetry and tensile in nature due to the hydrostatic pressure being positive. The radial stress is negligible compared to the tangential and meridional stresses and a plane-stress condition can be approximated. The wall thickness is small compared to the diameter of the sphere, hence, all stresses are considered to be a constant through the wall thickness. As such, no bending or torsional stresses are experienced in the metal.

Thick-walled spheres have a wall thickness which is not small compared to the diameter of the sphere. The stresses are a function of radius through the wall and the radial component of stress can be a significant quantity. As such, there exists a triaxial stress-state in the metal which can give rise to significant bending stresses and associated decreased mechanical stability i.e., buckling can occur more readily.

Although Figure 1 shows the D/t_{metal} ratio for the microsphere to be borderline between thin and thick-walled conditions, analysis of test results for a thick-walled condition are unreliable as compared to a thin-walled approximation. The triaxiality of the stress-state under thick-walled conditions presents an extremely difficult stress-strain analysis as well as the aforementioned mechanical instabilities. Another

problem with a thick-walled condition is that a prohibitively large amount of material must be deposited onto a substrate. For these reasons a thin-walled test set-up will be used to insure a biaxial stress-state in the material. Figure 2 illustrates these stress conditions and shows Mohr's circle of stress for a thin-walled spherical shell under internal pressure.

Experimental Procedures

Numerous methods exist for creating a biaxial stress-state in a relatively thin sheet of metal. These include clamping down relatively thick sheets of metal as is done in deep drawing experiments to obtain forming limit diagrams,⁽³⁾ measuring limit strains by changing an imposed strain ratio, $\rho = \epsilon_2/\epsilon_1$, in the biaxial stretching of thin sheets of metal,⁽⁴⁾ and clamping down a thin metal film and deforming it using a solid hemisphere⁽⁵⁾ or fluid pressure.⁽⁶⁾ The mechanical test which appears best suited to the actual in-service conditions is the bulge test using fluid pressure, in which a thin metal film is clamped over a circular cylinder and subjected to a controlled hydrostatic pressure on one side. The resulting deflection, or bulge, of the film is measured at the pole of the film as a function of applied pressure. In this manner an equivalent stress-equivalent strain curve is generated for small deflections and may be compared with equivalent stress-equivalent strain curves for other materials and processes.

Tsakalakos⁽⁷⁾ has developed a reliable bulge test set-up for measuring strains up to about 1%. He used this procedure to measure the biaxial elastic modulus from the slope of stress-strain curves for certain crystallographic orientations of a Cu-Ni alloy and found the measured values to be consistently in agreement with calculated values from the bulk alloy elastic constants. The basic procedure is to deposit the metal atoms onto the desired substrate until a film of the desired thickness and size is obtained. The film is then peeled off the substrate while submerged in water and floated back onto the substrate as flat as possible. The film is then allowed to air dry. A rigid metal frame with a hole in the center is placed over the film and attached to it using scotch tape. A washer with well defined edges is then glued to the film using a room temperature curing epoxy, hence, avoiding the introduction of residual stresses into the film. The film is then cut around the outside diameter of the washer and placed film side down into the bulge tester. The inner diameter of the washer will become the bulge aperture. Figure 3 is a schematic diagram of the test set-up to include bulge tester, laser interferometer and viewing screen. Interference occurs between the pole of the developing bulge and the bottom of a vertically adjustable glass slide, not shown in the figure. The glass slide is above the film and is adjusted such that at zero film deflection a minima occurs at the center of the viewing screen. As the film bulges outward fringes are created and each new fringe corresponds to a deflection of the pole of the film of $\lambda/2$, where λ is the wavelength of light used. An upward bulge causes the fringes to move radially outward

on the viewing screen while a downward bulge causes the fringes to move inward.

Stress and Strain Analysis

Figure 4 indicates the method used to calculate stresses and strains from the measured quantities P , w_0 , a , and t , where, P is the difference in pressure between the two sides of the film of thickness t , w_0 is the height of the center of the bulge measured from the initial film plane, and a is the radius of the circular aperture which defines the initial test area. Stresses and strains are determined on the assumptions that the bulge is a cap on a sphere and that there is a uniform strain distribution in the film. Various researchers^(7,8,9,10,11) have shown these assumptions to be approximate at best. In fact, the geometry of the developing bulge is initially elliptical, progresses to hemispherical, and finally becomes parabolic, so that the radius of curvature at the pole of the bulge could more closely be approximated by a surface of revolution of a general quadratic whose form could be obtained by an analysis of two deflections, i.e., the central deflection and a quarter-chord deflection.

A mathematically rigorous derivation of strain⁽¹²⁾ indicates the strain to vary from zero at the clamped edges, $r = a$, to a maximum at the center of the film, $r = 0$. Hence, the value of the constant, f , in the strain equation is approximate, whereas the value of h in the stress

equation is more exact. Papirno tested thin foils of annealed aluminum and measured the polar strain directly using strain gauges. He obtained the empirical relation $\epsilon = 0.30 (w_0/a)^{1.9}$ by sighting the best straight line through the data graphed logarithmically. If one assumes a biaxial stress-state at the pole of the film, then the linear portion of the stress-strain curve may be expressed using Beams' solution, i.e.,

$$\sigma = \sigma_0 + \frac{2}{3} \left(\frac{E}{1-\nu} \right) \left(w_0/a \right)^2, \text{ in which,}$$

$$\epsilon = \frac{2}{3} \left(w_0/a \right)^2, \text{ where,}$$

E = Young's modulus, ν = Poisson's ratio, and σ_0 = a constant which depends on the stress-state in the film at zero deflection. The value of σ_0 is obtained from the initial slope of the P vs. w_0 curve, whereas Young's modulus is obtained from the elastic portion of either the

$$\Delta P \text{ vs. } (w_0/a)^3 \text{ curve, where } P_0 = \frac{4t w_0^3 \sigma_0}{a^2},$$

or from the stress-strain curve plotted as P/w_0 vs. $(w_0/a)^2$.

Reported values of Young's modulus^(6,10) and tensile strength⁽⁶⁾ for thin films are larger than the bulk values and indicate these increase with decreasing film thickness, for film thickness below about 5000Å. The possible strengthening mechanisms involved are discussed by Mentor and Pashley.⁽¹³⁾

Figure 5 is an example calculation showing the applicability of the bulge test to the service conditions experienced by the filled microspheres. For typical values of P , w_0 , a and t , the stresses in the bulged film will be equal to or greater than those experienced by the metal coating on the microspheres, hence, a quantitative analysis will indicate which materials have mechanical properties most desirable in containing the high pressure gas.

Mechanical Aspects of Bulge Testing

To insure a biaxial stress-state in the film the ratio, a/t must be greater than about 10^3 , hence, if $a = 5 \text{ mm}$ then $t \leq 5 \text{ } \mu\text{m}$. Some problem may exist in defining a , the radius of the circular aperture. Usually, it is assumed that the only material which contributes to the overall deformation, elastic or plastic, is the volume encompassed by $r \leq a$. If some slippage occurs at the clamped ends, or if some of the metal under the clamp (washer) deforms, the measured displacement may not be indicative of material properties. This can be corrected for by considering a to be an effective circular aperture, i.e., a_{eff} , calculated by considering the final-plastic-film thickness and assuming a linear dependence of the effective circular aperture. Although this procedure involves the stiffness of the testing machine, i.e., deflection of the washer, preliminary calculations show this quantity to be small. Another problem is in the definition of $w_0 = 0$, that is, if the film is wrinkled to begin with the actual onset of material deformation will be delayed.

The solution is to plot P vs. w_0 and extrapolate to $P = 0$ on the w_0 axis.

Circumventing the uncertainty in the stress-strain equations may be accomplished by utilizing noncircular apertures, i.e., rectangular holes. In this manner, the bulged surface can be considered a section of an internally-pressurized thin-walled cylinder for which exact stress-strain solutions exist. However, Beams compared results obtained by bulging gold and silver films through rectangular holes, 3 mm x 15 mm, to results obtained by bulging through circular apertures, $2a \approx 3\text{mm}$, and found no significant differences.

The advantages of testing thin films using a bulge method are numerous. The usual difficulties encountered in a uniaxial tension or compression test, such as, specimen preparation, mechanical alignment and possible tearing of thin films are greatly reduced. Deforming the metal film using gas pressure, instead of a solid hemisphere or oil pressure, alleviates frictional effects and the possibility of oil spillage, respectively.

Metallurgical Aspects in Bulge Testing

We would like to be able to compare the equivalent stress-equivalent strain curves from the bulge test data to results obtained from a uniaxial compression test, the equal biaxial stretching and associated thinning in

the radial direction of the film being equivalent to compressing the material. However, performing a compression test on a thin film is unrealistic, as such, uniaxial tension results are the next best thing. The problem with comparing tension test results to bulge test results is that the concept of equivalent stress-equivalent strain is based on the assumption that the flow stress, at a given strain, is not a function of the stress-state in the material. That is, all the yield criteria (Von Mises, Tresca, etc.) assume that plastic flow in the material is the result of a shear stress only. If this were the case, the same true stress-true strain curve would result when the same material was tested in tension or compression, the only difference between the two being the sign of the hydrostatic component of stress. This has been found not to be the case by many researchers.⁽¹⁴⁾ The well known strength-differential effect,^(14,15) defined as a difference in the flow stress between tension and compression, at a given strain, can be explained in terms of an interaction of the rate-controlling activated configuration with the hydrostatic component of the applied stress. Therefore, performing a bulge test on thin films of metal and obtaining an equivalent stress-equivalent strain curve may be a relative indication of the mechanical properties of the material, it should not be taken as necessarily representative of an uniaxial tension or compression test. There may be some effect of the planar-hydrostatic component of stress on the resulting flow behavior of the material.

The limit strain, defined as the total uniform strain in the material, will be controlled by either the strain to fracture or the strain prior to the onset of a localized plastic instability. The fracture strain is very sensitive to the positive-planar-hydrostatic component of stress, whereas, in equibiaxial tension the development of a local strain inhomogeneity is inhibited and the material develops diffuse necking. Eventually, a strain inhomogeneity will occur in the form of a narrow localized neck followed soon by fracture. Most workers⁽¹³⁾ have found relatively little plastic deformation prior to fracture, hence, the total uniform strain in a hydraulic bulge test tends to be less than the total uniform strain observed in uniaxial tension or compression tests. Surface irregularities, loading rates, initial internal stresses, local grain size fluctuations and grain shape or crystallographic anisotropy may play a significant role in this process. Wilson, et al⁽¹⁶⁾ performed bulge tests on sheets of copper, Cu-30 Zn brass and low-carbon steels without strongly developed preferred orientations and found the limit strains decreased with decreasing t_0/d_0 , where d_0 is the average grain diameter and t_0 the sheet thickness. For $0.4 \text{ mm} \leq t_0 \leq 1.2 \text{ mm}$, he concluded that when t_0/d_0 was less than about 20, plastic anisotropy of individual grains of the primary phase was the dominant source of strain inhomogeneities leading to eventual necking failure. Wilson⁽¹⁷⁾ also tested highly textured sheets of copper, aluminum and low carbon steels and found the limiting strains, in equibiaxial tension, to be significantly lower than those of weakly textured sheets with similar t_0/d_0 ratios.

Mohr's circle of stress, Figure 2, indicates that, for the radial component of stress equal to zero, the maximum shear stress occurs at 45° to the principal stress direction. Since the radial component of stress is not zero, but varies from $\sigma_r = P$ at the inside surface, to $\sigma_r = 0$ at the outside surface, the direction of the maximum shear stress will be a function of radius through the thickness. A slip-line field theory analysis should yield this dependence. In either case, the degree of preferred orientation in the films should be obtained to determine the relationship of crystal orientation to the principal stress axis as this will effect the flow behavior of the material. Local grain size fluctuations and grain shape anisotropy may also influence the mechanical properties of thin films. The Hall-Petch⁽¹⁸⁾ relation, $\sigma_t = \sigma_0 + kD^{-1/2}$, where, σ_0 = the yield stress, σ_i = a friction stress opposing dislocation motion, k = a constant and D = the grain diameter, expresses the grain-size dependence of the flow stress for a given plastic strain, and states an increased flow stress will result as the grain size decreases, corresponding to an increased grain boundary area per unit volume of material. One interpretation of this model states that grain boundaries can act as barriers to dislocation motion, hence, if the vapor-deposited thin film exhibits a void-columnar grain structure (aspect ratio $\neq 1$) the resulting flow behavior of the material will differ from a thin film with small equi-axed grains.

Suggestions for Future Research

The mechanical properties of thin films need to be measured using a hydraulic bulge test method and compared to uniaxial tensile results. Care must be taken in the interpretation and analysis of these test results as outlined in this paper. X-ray diffraction experiments should be performed on the thin films to determine the degree of crystallographic anisotropy, as well as the usual metallographic analysis to evaluate grain size fluctuations and grain shape anisotropy. Stress relaxation measurements should be performed as this method will yield more reliable values for the bulk modulus. Films should be tested to failure and fracture surfaces analyzed using SEM techniques to determine the mode of fracture.

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FIGURE 1

INITIAL TARGET DESIGN

$$100 \text{ atm} \leq P_{\text{D-T}} \leq 200 \text{ atm}$$

$$D_i / t_{\text{METAL}} = 20:1$$

EXAMPLE COMPOSITE TARGET

FIGURE 2

THIN-WALL

THICK-WALL

IF: $D/t \geq 20$

USE MEMBRANE THEORY

$D/t < 20$

USE THICK-WALL THEORY

MEMBRANE THEORY

σ_θ = TANGENTIAL (HOOP) STRESS $\neq f(r)$

σ_ϕ = MERIDIONAL STRESS $\neq f(r)$

$\sigma_\theta = \sigma_\phi$ $\sigma_r \ll \sigma_\theta, \sigma_\phi$

FIGURE 3

BULGE TESTER

* SPHERICAL APPROXIMATION
SEE FIGURE 4

FIGURE 4

W_0 = bulge height
 a = radius of aperture
 r = radius of equivalent sphere

$$W_0 = r(1 - \cos \theta)$$

$$a = r \sin \theta$$

STRESS:

$$\sigma_\theta = \sigma_\phi = \sigma = \frac{Pr}{2t} = \frac{Pa^2}{W_0 t} \underbrace{\left[\frac{1}{2} \left(\frac{1 - \cos \theta}{\sin^2 \theta} \right) \right]}_h ; \quad \lim_{\theta \rightarrow 0} h = \frac{1}{4}$$

$$\therefore \underline{\underline{\sigma = \frac{Pa^2}{4W_0 t}}}$$

STRAIN: ($\epsilon \leq 10\%$, $\therefore \epsilon = E$)

$$\epsilon = \Delta l / l_0 = l_F / l_0 - 1 ; \quad l_F = r \theta, \quad l_0 = a$$

$$\therefore \epsilon = \frac{r \theta}{a} - 1 = \frac{W_0^2}{a^2} \underbrace{\left[\frac{\theta \sin \theta - \sin^2 \theta}{(1 - \cos \theta)^2} \right]}_f ; \quad \lim_{\theta \rightarrow 0} f = \frac{2}{3}$$

$$\therefore \underline{\underline{\epsilon = \frac{2}{3} W_0^2 / a^2}}}$$

DERIVATION OF STRESS AND STRAIN

FIGURE 5

a) D-T FILLED MICROSPHERE -

FOR: $D_i = 200 \mu\text{m}$, $P = 200 \text{atms.}$, $t_{\text{METAL}} = 10 \mu\text{m}$

$$\sigma_\theta = \sigma_\phi = \frac{Pr}{2t} = (200 \text{atms})(100 \mu\text{m}) / 2(10 \mu\text{m}) = 1000 \text{atms} \\ = 15 \text{Ksi}$$

b) BULGE TEST -

FOR: $P = 10 \text{psi}$, $a = 5 \text{mm}$, $t = 5 \mu\text{m}$, $w_0 = 0.5 \text{mm}$

$$\sigma = \frac{Pa^2}{4w_0t} = (10 \text{psi})(5 \text{mm})^2 / 4(0.5 \text{mm})(5 \mu\text{m}) \approx 1667 \text{atms} \\ = 25 \text{Ksi}$$

THIS STRESS CORRESPONDS TO A STRAIN OF ABOUT 1%.

COMPARISON OF STRESSES IN A D-T FILLED MICROSPHERE TO
THOSE IN A BULGED THIN METALLIC FILM