GA–A24458

FABRICATION OF GAS-FILLED TUNGSTEN-COATED GLASS SHELLS

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SEPTEMBER 2003

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This is a preprint of a paper presented at the 15th Target Fabricartion Specialists Meeting, Gleneden Beach, Oregon, June 1–5, 2003 and to be published in *Fusion Science and Technology.*

Work supported by
the U.S. Department of Energy
under Contract No. DE-AC03-01SF22260

GENERAL ATOMICS PROJECT 30095
SEPTEMBER 2003

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Deuterium (D_2) filled glass shells coated with a high Z element are needed for high energy density (HED) experiments by researchers at Los Alamos National Laboratory. We report here on our initial attempt to produce such shells. Glass shells made using the drop tower technique were coated with gold, palladium or tungsten, or a mixture of two of these elements. It was found that gold and palladium coatings did not stick well to the glass and resulted in poor or delaminated films. Tungsten coatings resulted in films suitable for these targets. Bouncing of shells during coating resulted in uniform tungsten coatings, but the surface of such coatings were filled with small nodules. Proper agitation of shells using a tapping technique resulted in smooth films with minimal particulate contamination. For coating rates of ~ 0.15 μm/hr coatings with ~ 2 nm RMS surface finish could be deposited. The surface roughness of coatings at higher rates, 0.7 μm/hr, was considerably worse (~ 100 nm RMS). The columnar structure of the coatings allowed permeation filling of the tungsten coated glass shells with deuterium at 300°C.

I. INTRODUCTION

Deposition of uniform high Z coatings on shells have been investigated by magnetron sputtering previously by others for applications in the Inertial Confinement Fusion (ICF)[1,2] and Inertial Fusion Energy (IFE)[3] areas. In particular, our group has deposited gold and palladium on polymer shells for IFE applications. In this report we present our initial findings on deposition of gold, palladium and tungsten on glass shells for HED applications.

This task presented several challenges. The coatings were required to be ~ 3 μm thick and uniform. Therefore, stock mounting of shells was not a viable option due to the inherent non-uniformity introduced by that technique. This necessitated deposition while shells were being agitated and solving the associated problems as described later. In addition to the coating, the glass shells needed to contain ~ 10 atm of D_2 gas. Permeation filling of D_2 into uncoated glass shells is possible in a reasonable time at ~ 360°C. However, it was not clear apriori whether the glass shells should be filled before or after the coating. Permeation filling through the ~ 3 μm of deposited coating was an untried possibility. Equivalently, it was not clear whether the glass shells would retain the fill during the deposition process if pre-filled prior to coating. As described below, we have made significant progress in answering many of these questions through this initial investigation.

II. EXPERIMENTAL DETAILS

Gold, palladium and tungsten were deposited using magnetron sputtering. The glass shells were required to be ~ 850 μm in diameter and ~2–3 μm in thickness. They were made using a heated drop tower. The glass shells were agitated in two different ways. In the first technique, the shells were loaded into a "bounce" pan and the pan was shaken at ~ 100 Hz using an electromagnetic shaker. In the second scheme, the shells were loaded into a container and an electromagnetic solenoid was used to tap the container once every 5–10 s. Since the substrates were glass they could be placed relatively close to the magnetron gun, as close as ~ 50 mm without being affected by the heat generated (> 100°C) by the coating process. However, the source to substrate distance was varied between ~50–125 mm. The coating rate was ~0.15–0.7 μm/hr depending on the proximity of the shells to the gun. Typically 10–20 shells could be coated at once regardless of the agitation scheme used.

Coating thickness was measured by several techniques. Shells were weighed before and after the coating with ~ 0.3 μg accuracy to determine the average coating thickness assuming full density coatings. 1 μm of tungsten coating weighs about 40 μg at this diameter. Therefore, this offers an accurate non-destructive average thickness measurement technique. However, the density of the coating is not necessarily the same as the bulk

material. Therefore, the coatings were also measured by interferometry by destroying witness shells from the run. Finally, in some cases coated shells were examined by scanning electron microscopy (SEM). The coating could be measured to better than ~ 0.3 μm using these techniques. In addition, flat witness plates positioned near the shells were included in each run to get further indication of the coating thickness, although indirectly in this case. The uniformity of the coatings was measured mainly by rolling shells down an incline[4] and noting the tack angle they took. Also, a number of shards obtained from destroying the witness shells were measured to obtain further uniformity data. Glass shells were permeation filled with ~ 10 atm of D_2 at ~ 300°C in about a day for experiments involving the gas fill. Coating surface roughness was measured by atomic force microscopy (AFM) and by phase shift interferometry (PSI).

III. RESULTS AND DISCUSSION

Initially, gold coatings were attempted. The glass shells were agitated using the bouncing technique. They could be sufficiently agitated prior to turning on the magnetron. However, within minutes of commencing the gold deposition, the shells would stick to the bounce pan surface and stop moving. The shells could be jarred loose and forced to move by continuously tapping against the bounce pan. While the deposited gold appeared rather shiny and uniform visually, microscopic examination showed major tearing of the coating at numerous spots around the shell as evidenced in the SEM image shown in Fig. 1. The coating thickness is ~ 0.5 μm. This is similar to what has been observed in previous reports.[1,2] Tearing of the gold coating had also been observed in our previous work on coating of polymer shells but to a lesser degree.[3] Since coating palladium had resulted in less delamination from polymer shells in our previous experience, we decided to try it on glass shells as well. While the results with palladium were better and less sticking was encountered in agitating the glass shells, there was still serious tearing of the deposited film from the glass shells (Fig. 2).

We then decided to deposit tungsten, since it is a high Z element and it is not a noble metal as in the case of gold and palladium. The hope was that tungsten would not exhibit the "cold welding" associated with gold and palladium coatings. The behavior of the glass shells in the bounce pan was markedly different when tungsten was deposited. The sticking of shells to the bounce pan observed in coating gold and palladium was conspicuously absent. The coatings looked very shiny and microscopic examination revealed a continuous coating with no observable tearing. However, as shown in the SEM image of Fig. 3(a), the surface finish of these shells was far from smooth even for coatings of ~ 1 μm thick. The RMS surface roughness range was typically in the few hundred nm range. The surfaces of shells in these coatings were cluttered with small nodular growths which were not present on flat substrates coated along side the shells as witness samples. This indicated that the nodules were not related to the sputter deposition itself, but rather associated with coating on shells. The recent work on Beryllium sputter coatings on shells by McEachern, et al.[5] has pointed out that coatings on shells can be much rougher than those on flats.

While there was no stringent surface finish requirement (eventually better than ~ 100 nm RMS is required) on these shells, we decided to change the agitation mechanism to examine possibly obtaining smoother coatings. The shells were placed in small containers and agitated by continuously tapping the container. The containers were chosen to be only about twice the shell diameter, small enough to prevent self shadowing effects as well.[5] This arrangement resulted in very smooth coatings. Figure 3(b) shows an SEM image

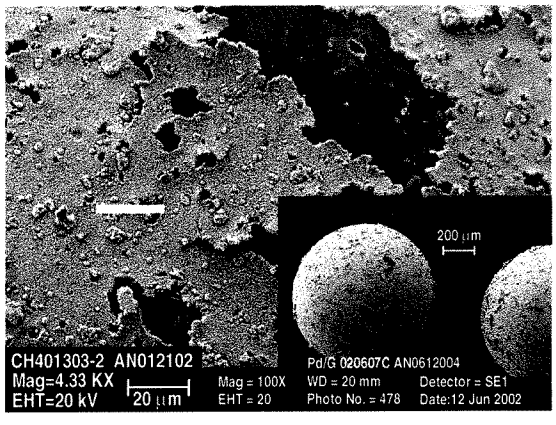

Fig. 1. SEM image of ~ 0.5 μm gold coating on glass shells. The shells were forced to move during the run by tapping on the bounce pan. Major tearing of the gold coating from the shells is seen. The inset shows the full shell. Bar is 20 μm.

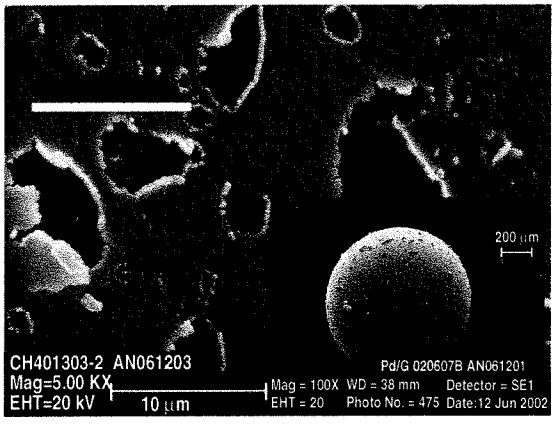

Fig. 2. SEM image of ~ 0.5 μm palladium coating on glass shells. The shells were forced to move during the run by tapping on the bounce pan. Tearing of the palladium coating from the shells is seen, although it is less than what was observed with gold shells. The inset shows the full shell. Bar is 10 μm.

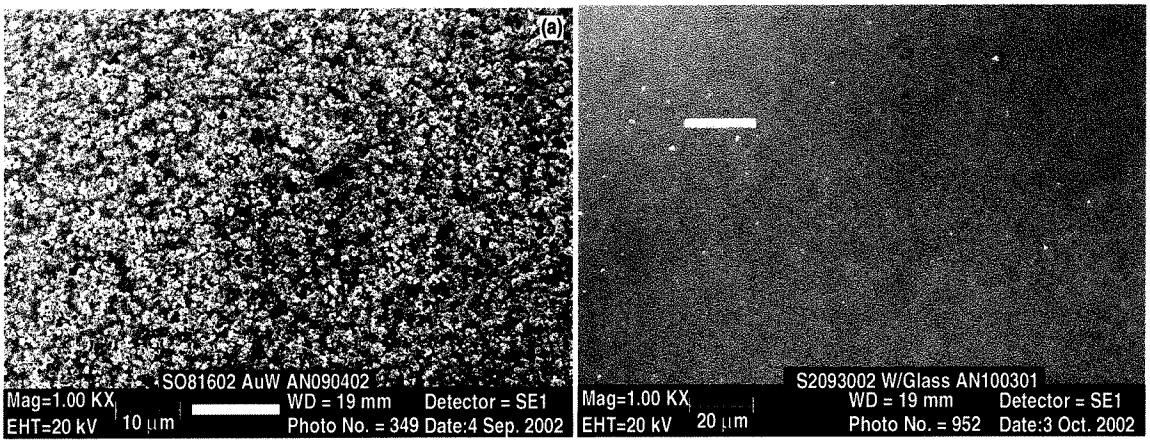

Fig. 3. (a) SEM image of ~ 1 μm coating of W on a glass shell. The shell was bounced coated. There is little evidence of tearing of the coating away from the shell, but the coating is rough due to growth of features. The coating rate was ~ 0.15 μm/hr. Bar is 20 μm. (b) SEM image of ~1 μm coating of W on a glass shell. This shell was coated using tapping agitation. The surface is remarkably smooth compared to the bounce coated shell of Fig. 3(a). The coating rate was the same as in (a). Bar is 20 μm.

of the surface of a shell coated with ~1 μm of tungsten using this agitation scheme. The improvement in the surface finish is rather remarkable. AFM patch scan measurement of the surface is shown in Fig. 4. The RMS surface roughness was only 2 nm. It should be noted that the substrates were ~ 125 mm away from the gun in these coatings, resulting in a rather low deposition rate of ~ 0.15 μm/hr.

Given this marked improvement in the surface finish, the tapping scheme was used in all subsequent runs.

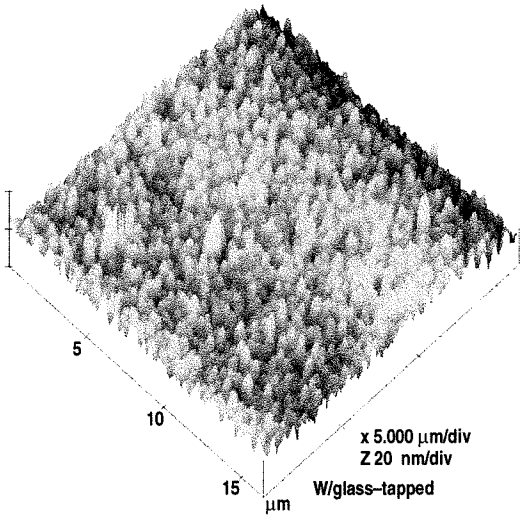

Fig. 4. AFM image of coating shown in Fig. 3(b). The RMS surface roughness is only 2 nm.

Coatings as thick as ~ 4 μm were deposited. However, due to the desire to increase the coating rate, the shells were moved close to the gun to within a ~ 50 mm separation. This resulted in increased coating rate of ~ 0.7 μm/hr which allowed faster processing of batches. Many batches of shells were coated to between 3 and 4 μm in thickness to verify the reproducibility of the coating process which is ultimately essential for reliable production of these targets. The coating runs were indeed very reproducible and no major variation was observed from run to run. Also the yield of coated shells from a given run was 90% or better indicating a low attrition rate for the process. The surface finish of the coatings at this higher coating rate and for these thicker coatings increased substantially to ~ 90 nm RMS. This was still within the range desired for this initial work.

The microstructure of the coatings under these conditions was examined in cross section by SEM on a number of samples. An example is shown in Fig. 5 for a ~ 3 μm tungsten coating. The coatings exhibited a columnar structure as seen in the image. The density of the coatings was measured by comparing the thickness obtained by weighing to that obtained by interferometry. The inferred density was only ~ 80% of the bulk density. This is surprising given that the SEM images do not show any major voids in the coatings. The major error in this density measurement is in the measurement of the wall thickness by interferometry which is ~ 15%. This discrepancy in the thickness measurements and its apparent implication on the low inferred density of the coating is puzzling and will be examined further.

Having established reproducibility in the coating step we next began a preliminary examination of the issue of D_2 filling the shells. Initially, the glass shells were filled with D_2 prior to the coating run. The coating was performed with the shells only 50 mm away from the gun as before. After the run, a few shells were broken and it

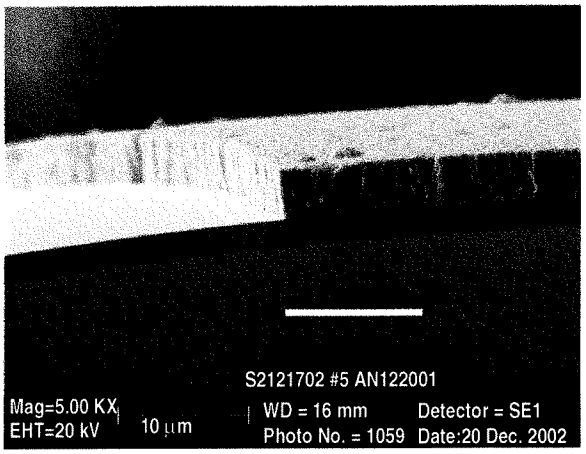

Fig. 5. SEM image of ~ 3 μm coating of W on a glass shell. This shell was coated using tapping agitation. The coating rate was ~ 0.7 μm/hr. The familiar columnar structure seen in sputter coatings is visible.

was found that only ~ 3 atm of D_2 was left inside the shell after the run. The glass shells had apparently gotten hot enough for the D_2 to escape. This is again surprising as D_2 permeation through glass is rather slow unless the temperature is quite high (> 300°C). A heat sensitive tape placed in the proximity of shells confirmed that the temperature of the shells was ~ 300°C during the coating. Furthermore, D_2 must have permeated through the coated tungsten layer as well since loss of ~ 70% of the D_2 through glass should have taken place on the order of hours, during which time a substantial amount of tungsten was deposited on the shell. It was therefore conjectured that the coating itself might also be permeable to D_2 at elevated temperatures. Initial tests have proven this to be the case. Several shells coated with ~ 3 μm of tungsten were indeed successfully permeation filled with D_2. The fill was confirmed by breaking the shells as before. Therefore, this provides a path for fabrication of the desired targets. The room temperature permeation of glass is low enough that after permeation filling of tungsten coated shells at ~ 300°C, the gas is trapped inside the shell at room temperature.

In future work, more experiments need to be performed to verify the reproducibility of permeation filling through tungsten coated glass shells. Also, the possibility of filling glass shells prior to the coating and then coating at larger source-to-substrate distances (keeping shells cooler) to reduce or potentially eliminate loss of D_2 fill from shells during coating needs to be investigated. Larger source-to-substrate distances may eventually be preferred because they result in better surface finishes as well. The lower coating rate in that case is not a major issue in principle and longer coating runs will be required. It will also be interesting to reexamine gold or palladium coatings using the new agitation scheme to see if the problems encountered in the bounce pan technique are reduced with the tapping technique.

IV. RESULTS AND DISCUSSION

The initial effort for fabricating D_2 filled glass shells coated with tungsten for HED experiments by researchers at Los Alamos National Laboratory has been successful. Gold and palladium coatings did not stick well to the glass and resulted in poor or delaminated films. Tungsten coatings were continuous and adherent to the glass substrate. Bouncing of shells during tungsten coating resulted in uniform tungsten coatings, but the surface of such coatings were filled with small nodules. Proper agitation of shells using a tapping technique resulted in smooth films with minimal particulate contamination. For coating rates of ~ 0.15 μm/hr coatings with ~ 2 nm RMS surface finish could be deposited. The surface roughness of coatings at higher rates was considerably worse (~ 100 nm RMS). Tungsten-glass shells were permeation filled with D_2 at 300C. Further work needs to be done to verify the reproducibility of these results.

ACKNOWLEDGMENT

Work supported by the U.S. Department of Energy under Contract DE-AC03-01SF22260.

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