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OPTIMIZING HIGH Z COATINGS FOR IFE SHELLS

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ABSTRACT

Inertial Fusion Energy (IFE) reactors require shells with a high-Z coating that is both permeable and reflective. Previously gold was deposited on shells while they were agitated to obtain uniform, reproducible coatings [1]. However, these coatings were rather impermeable. We have coated Pd on shells in the same manner and used an x-ray fluorescence technique to accurately measure thicknesses and uniformities of the deposited layers on shells. We have demonstrated that these palladium coated shells are substantially more permeable than gold. Pd coatings on shells remained stable on exposure to deuterium. Pd coatings had lower reflectivity compared to gold that leads to a lower working temperature, and efficiency, of the proposed fusion reactor. Seeking to combine the permeability of Pd coatings and high reflectivity of gold, AuPd alloy coatings were produced using a co-sputtering technique. These alloys demonstrated higher permeability than Au, and higher reflectivity than Pd. However, these coatings were still less reflective than the gold coatings. To improve the permeability of gold's coatings, permeation experiments were performed at higher temperatures. With the parameters of composition, thickness, and temperature, we have the ability to comply with a large target design window.

I. INTRODUCTION

Fusion promises to offer a clean, inexpensive, efficient, and abundant source of energy for the future. Inertial Fusion Energy (IFE) [2], which involves compression of a small (≈ 4 mm in diameter) deuterium-tritium (DT) filled shell, or the target, by a driver such as a high power laser, is an alternative to the more conventional magnetic fusion scheme. Fabrication of the target for IFE experiments is a crucial step in making this scheme viable. The most promising current designs for IFE include a so-called radiation pre-heat direct drive target [3]. High Z material is needed to improve the implosion and lead to more energy release, or yield, from the target. The targets are to be injected in a hot reactor environment and the DT fuel ice layer will be exposed to excessive black body radiative heating which can cause it to melt, rendering the target useless. By choosing a metal with high reflectivity as the high Z material and concentrating it into an over-coating layer, this high Z material can protect the DT layer by reflecting radiation away. Low reflectivity of such a coating would require lowering the reactor temperature, which reduces the efficiency of conversion of released nuclear energy to electricity. Therefore, coatings with high reflectivities are desired. Due to the hazards of tritium, its degradation into He3 creating bubbles within the deuterium ice layer, and the practical functioning of a power plant (shooting ≈ 6 shells per second), the fill time must be short and the tritium inventory must be kept low. Therefore, the high Z coating must be permeable enough to deuterium to allow rapid filling. Using the relation that the fill time is directly proportional to shell permeability [4], the shell must be quite permeable.

The work reported in this paper describes deposition and characterization palladium as the high Z material, initial work on deposition and characterization of AuPd alloy coatings, and experiments of gold permeability at elevated temperatures. Gold was first chosen due to design experiments and its excellent reflectivity in the region of interest (≈ 0.5–25 μm). Au coatings, however, proved not permeable enough for a practical power plant. We chose to investigate Pd because of its known high permeability to hydrogen and its isotopes [5]. Recent experiments at Nike have shown that Pd can also be used as the high Z coating [6]. Pd reflectivity is known to be lower than Au, which is what our work found. Therefore, Au/Pd alloys were investigated in hopes of high permeability and reflectivity. These coatings were still less reflective than the gold coatings, so Au permeability was tested at elevated temperatures in an effort to optimize permeability and reflectivity.

II. EXPERIMENTAL TECHNIQUES

II.A. Deposition

Pd coatings were sputtered onto targets [7,8]. Films were deposited at a pressure of Ar of 30 mTorr, a power of 150 W, and source to substrate distance of 90 mm. We used both flat targets, such as glass slides and Si wafers, and shells. In the future this coating will actually go onto a foam shell. Since foam shells are not yet available, we used full density polymer shells, PAMS (poly alpha-methyl styrene). In order to uniformly coat shells we used an electromagnetic shaker that rapidly bounced the shells. A number of different variables could be controlled in the deposition process. These included the sputter gun power, gun-substrate distance, deposition pressure, and shell agitation. The pressure and gun-substance distance is particularly important because the longer the path or the more particles in the path, the more likely it is that the sputtered Pd atoms will scatter and lose kinetic energy before arrival on the substrate. Therefore both higher pressure and longer gun-substrate distance result in more scattering and less dense coatings. There was a minimum gun-shell distance, since the polymer shells cannot be brought too close or they will melt. We used the same deposition conditions on flats and shells for consistency.

II.B. Thickness Measurement by X-ray Fluoresence (XRF)

Since the thickness of Pd needs to be a narrow range (≈ 300–1000 Å) it is crucial to measure the coating thickness accurately. In addition, we need to ascertain any Pd thickness non-uniformity around the shells. Coatings are traditionally measured using profilometry. Profilometry, while possible on flats, is impractical to use on shells. A witness plate can be used for each coating run alongside the shells [9]. However, there are a number of problems with that technique.

The amount of metal deposited onto a flat such as the witness plate is not equal to that deposited onto a shell. If there were no scattering one would expect an area on a witness plate to have four times the amount of coating than that of an area of shell. This is determined through simple geometry. A shell's area is four times that of a flat and since these shells are bouncing randomly the entire shell surface is coated. But because of scattering of sputtered material on the way to the shells, the actual factor can be different from the theoretical value of 4. Furthermore, by necessity the witness plate must be positioned away from where the shells are being coated and therefore the coating it receives is not fully representative of that deposited on the shells. In

addition, no thickness uniformity information on the shells can be obtained when using a witness plate.

XRF provides a relatively simple easy, non-destructive method to determine the amount of metal deposited. In this method the sample is excited by an x-ray beam and emits x-rays characteristic of the elements present in the sample. A portion of the x-rays is captured by a detector and the resulting peaks correspond to the amount of element present in the sample [10]. In measuring shells the beam hits two spots on the shells (Fig. 1). The shell is aligned so that one beam hits the South Pole and the other hits a spot on the outer circumference. The resulting signal includes the metal from both spots. By rotating the shell around the z-axis (keeping the South Pole spot stationary) we could vary one of the spots measured instead of two. Therefore wall thickness of individual spots on the shell can be measured providing much needed uniformity data. To make this method more accurate we plan on blocking the beam emitted by the spot at the South Pole so that only one spot will be measured. In this process the beams traverse the PAMS shell mandrel, but we found that there was no attenuation from the PAMS [11].

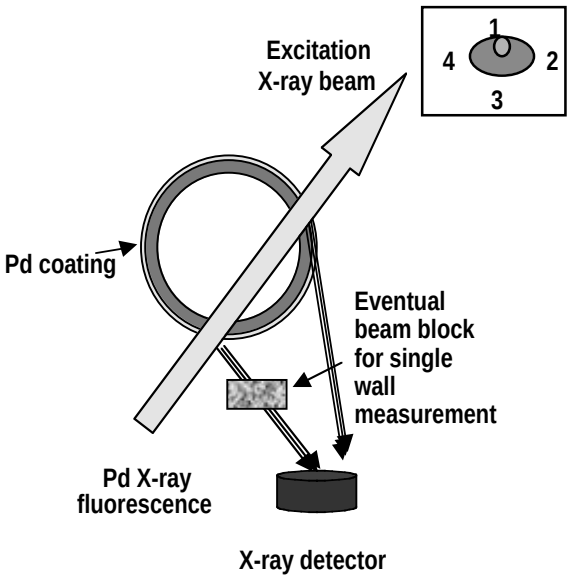

Fig. 1. Using XRF to determine the amount of metal deposited.

The composition was measured using energy dispersive x-ray analysis (EDX).

II.C. Permeability Measurements

In order to measure the permeability of our shells we placed the shell in a small volume chamber with a pressure gauge. After filling the chamber for several half-lives, such that the shell was nearly completely filled with gas, the chamber was evacuated leaving the filled shell in a small volume of low pressure. Then the pressure rise in the chamber resulting from the out-gassing of the shell was monitored. This rise in pressure was then fitted to a half-life. Background outgassing from gas absorbed in the chamber walls was taken into account when appropriate [12,13].

III. RESULTS AND DISCUSSION

III.A. Palladium

As mentioned previously, in an effort to improve permeability characteristics of the composite target, we turned to palladium to replace gold. Palladium has the unique quality of being highly permeable to hydrogen and so is frequently used as a catalyst and in purifying hydrogen. However, Pd films are also known to deform and crack greatly under exposure to hydrogen due to a large expansion of its crystal structure [14–18].

III.A.1. Characterization

We successfully coated polymer shells with palladium. We then analyzed the shells for uniformity and consistency, permeability, reflectivity, and cracking in the Pd layer upon exposure to deuterium. Uniformity measurements were performed using the XRF technique. By rotating the shell along the z-axis and keeping one spot fixed we could measure wall thickness at different points. The data from these measurements showed that the variations of wall thickness on an individual shell were within the measurement error [Fig. 2(a)]. We proceeded to check the shell to shell uniformity within a given batch. These measurements showed that the shells within a batch were uniform within the measurement error of about 10%. We then wanted to verify that shells of different batches were consistent [Fig. 2(b)]. In analyzing shells of different batches we also found consistency within 10%. So the shells produced are uniform and reproducible.

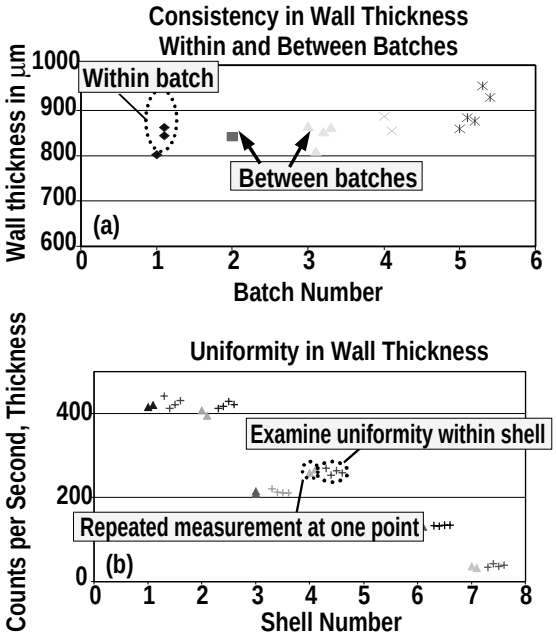

Fig. 2. (a) Variation of wall thickness of individual shells is within measurement error and (b) Different batches are consistent within measurement error.

III.A.2. Permeability

The attraction of palladium was its high permeability to hydrogen and its isotopes and our experiments confirm that. Due to radioactivity issues with DT, we used deuterium (D_2) gas as a

surrogate for the permeability measurements. Out-gassing data for deuterium through a bare polymer shell and a ≈ 1100 Å Pd coated polymer shell are very similar. In general, Palladium coated shells have a permeation time constant of about 10 ± 1 min, only slightly less permeable than the PAMS mandrel (≈ 8 ± 1 min). Therefore, Pd coatings are indeed very permeable when compared to gold coatings, which had time constants ≈ 1 hr for ≈ 250 Å coatings, as measured previously. Interestingly, we found that the permeability of palladium shells did not depend on the coating thickness. Shells of considerably different thickness exhibited the same time constant (Fig. 3). This implies that there may be a limiting step such as disassociation of the D_2 into palladium and then the gas can quickly pass through to the center. The variation of permeability between shells was within measurement errors, but any variation of the mandrels underneath the palladium would contribute to our measured variations in permeability of palladium shells.

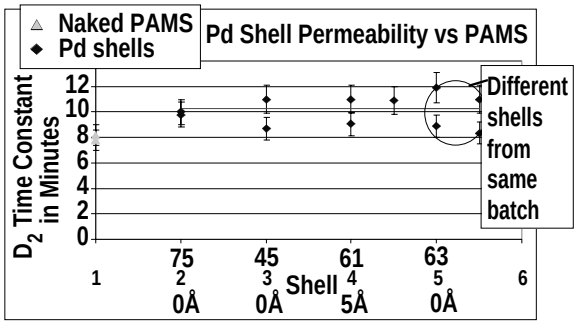

Fig. 3. Thickness of palladium coated shells does not affect permeability.

III.A.3. Deuterium Induced Deformation

In order to test for deformation we coated a glass plate with palladium and exposed it to D_2 for a period of several minutes. These plates were observed optically under a high power microscope during exposure. Immediately upon exposure the coating started to wrinkle and the deformities grew worse over time. When the D_2 exposure was terminated the coating began to lose its deformities, but the coating on the plate never returned to its original. In performing the same experiment with palladium coated shells we found no visual deformations (Fig. 4). In order to confirm that there were no microstructural changes unobservable under a microscope flats and shells were examined more closely. Examination using SEM confirmed these results; while the shells displayed the same amount of non-uniformity before and after exposure, the flats showed residual defects from D_2 exposure. Furthermore we saw no deformation in the shells that were repeatedly exposed to D_2 for longer periods of time in permeation measurements. From these results we infer that filling with DT should be rapid and cause no damage to the Pd coating layer.

III.A.4. Reflectivity

We expected the reflectivity of the palladium coatings to be lower, and this was confirmed. Although our palladium coatings had reflectivities that were close to the highest literature values for palladium, they were still below that of gold (Fig. 5). An important feature to notice is the large spread in the literature values. This demonstrates the effects of different deposition

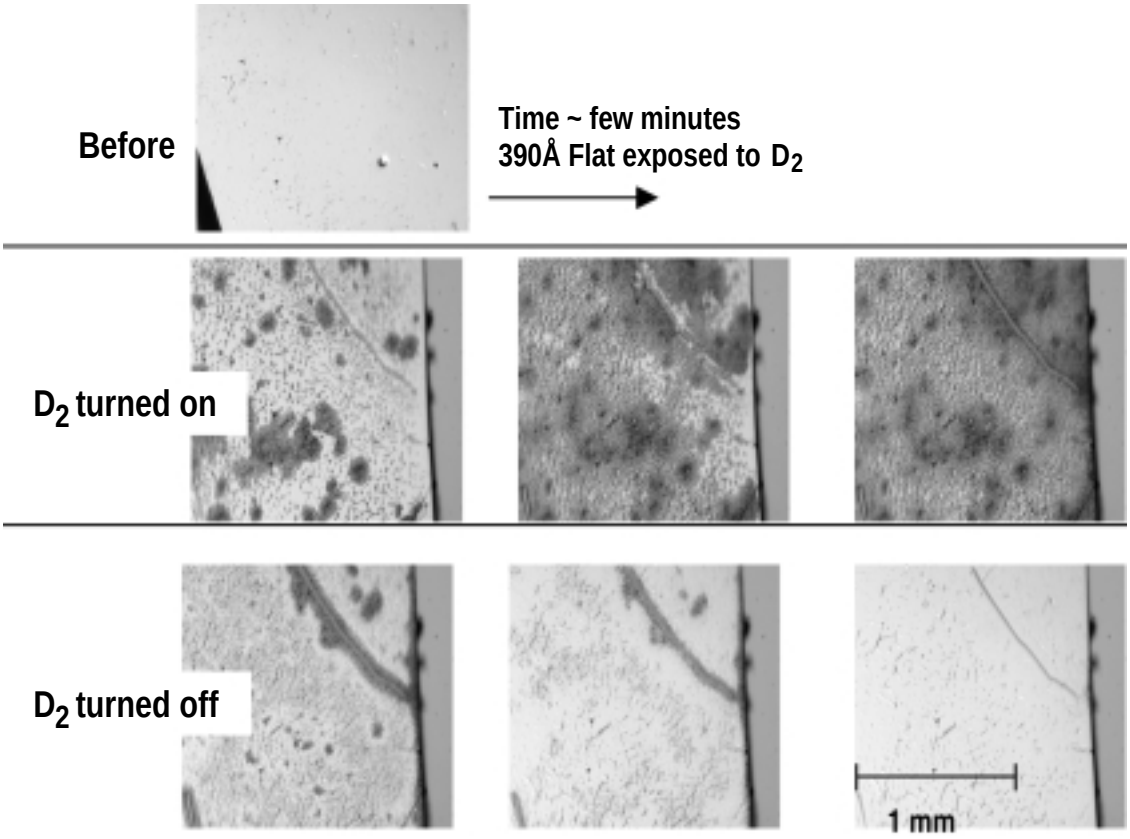

Fig. 4. Palladium coated shells exposed to D_2 did not deform.

conditions and measurement methods on reflectivity measurements [19]. Our measurements were done on flats using two independent methods, ellipsometry and direct measurement using an integration sphere. In ellipsometry, the materials optical constants, n and k values, are found which can then be used to determine reflectivity at arbitrary angles of incidence using the Fresnel reflection equations [20,21]. The second measurement is simply a direct measurement of the light reflected over all angles for a specific angle of incidence. It should be noted that reflectivities obtained by direct measurements were taken at discrete angles and were used for rapid feedback for process control and are adequate for comparisons only. Ellipsometry results provide reflectivities at all angles that are needed for calculation of the heat load on the target as it experiences the omnidirectional radiant heat while traversing the hot reactor chamber before being shot by the driver beams.

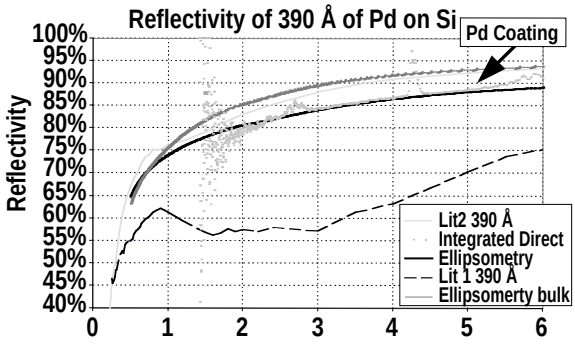

Fig. 5. Palladium coating reflectivities are close to highest literature values, but below that of gold.

III.A.5. Allowable Chamber Temperature

We convoluted the reflectivity data with a black body spectra for a given temperature to determine the total heat absorbed at various chamber temperatures. Then, by using the maximum heat absorption value for target survival determined by the detailed thermal calculations and simulations [22], we found the maximum chamber temperature for which shells coated by various thicknesses of Pd would survive the chamber heat (Fig. 6). The calculated values for gold coated (≈ 300 Å) shells are also shown. While the gold coated shells could survive at a maximum temperature of 1300 K, the palladium coated shells could survive at a maximum of 100 K. While this is a disadvantage of the Pd coatings, we next examined coatings involving mixtures of Pd and Au in hopes of obtaining coatings that have high permeabilities, due to Pd presence, and high reflectivities, due to Au presence.

III.B. Au/Pd Alloys

Initial analysis of AuPd alloys reveals that AuPd alloys coated shells have higher permeabilities than Au and higher reflectivities than Pd coated shells. AuPd alloys were successfully coated onto PAMS mandrels using co-sputtering technique. Two separate guns were used, one pure Pd and one pure Au, and an alloy was created during the sputtering process. In order to accommodate both guns, the guns were positioned at ~45° angle relative to the substrate, as opposed to 90° when sputtering a pure metal. Different compositions were made by varying the power of each gun. Layered coatings were also created in our attempt to create an optimal combination of reflectivity and permeability. As indicated in Fig. 7, the permeability of the alloy coatings were essentially equal to that of Pd. The reflectivities of these alloys were higher than the Pd values, although still below that of Au

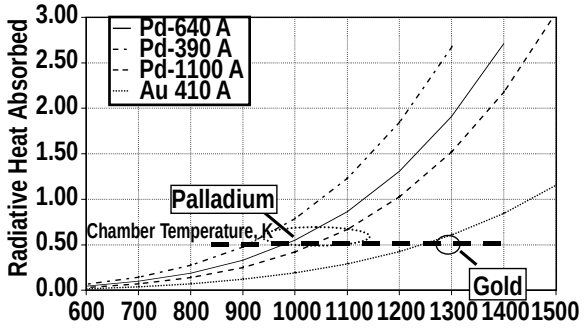

Fig. 6. Maximum survivable chamber temperature for coated shells.

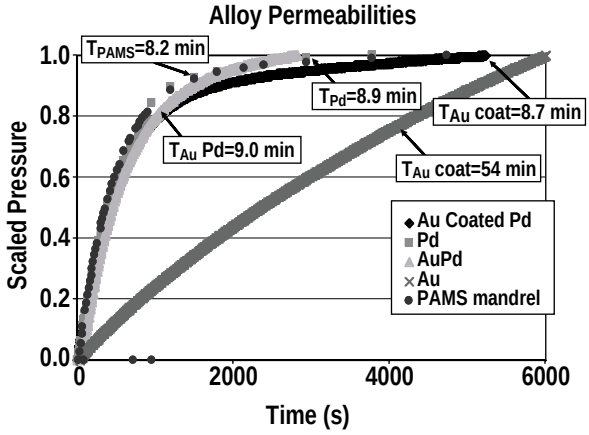

Fig. 7. Permeability of alloy coated shells is essentially equal to that of palladium.

(Fig. 8). A black body convolution was performed and the maximum chamber temperature was found to be higher than that of Pd, but lower than that of Au (Fig. 9). In plotting the percentage of gold versus reflectivity we found that deposition conditions significantly affect the reflectivity of the shell. This was apparent in comparing the reflectivity of a pure Pd coating in the new 45° set up compared to the reflectivity of a pure Pd in the 90° set up (Fig. 10). Due to this result we theorized that moving the Pd gun back to 90° while the gold remained at ~45° would improve the overall reflectivity. Gold did not appear to exhibit a difference in reflectivity at the ~45° angle as opposed to the 90° angle.

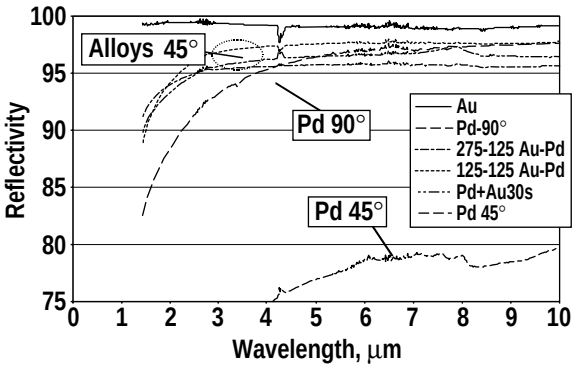

Fig. 8. Reflectivities of alloy coated shells are higher than palladium, but below that of gold.

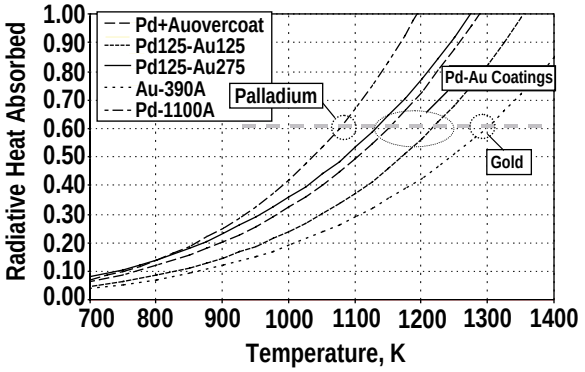

Fig. 9. Maximum survivable chamber temperature for alloys is higher than palladium but lower than gold.

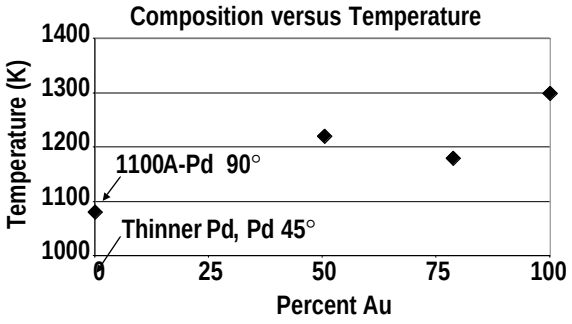

Fig. 10. Deposition conditions affect reflectivity.

V. TEMPERATURE VARIED PERMEATION

Another way to optimize the permeability and reflectivity of the high-Z coating is to improve gold's permeability by increasing the fill temperature. This was examined by performing Au fills at various temperatures. The same basic permeation set-up was used but with a bake-able baritron, and several thermocouplers to monitor temperature. Initial results (Fig. 11) indicate that Au coatings are about five times more permeable at 70°C than 21°C, and eight time more permeable at 105°C compared to 21°C. This is a vast improvement considering the PAMS permeability increased 5–6 fold from 21°C to 105°C. Looking at actual permeabilities a Au shell has a time constant of 375 s at 105°C compared to bare PAMS's time constant of 619 s.

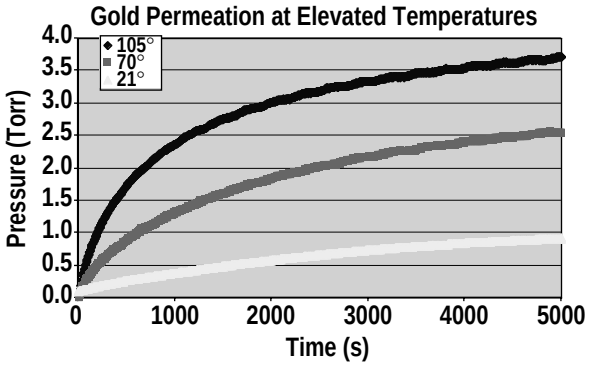

Fig. 11. Gold coated shells are ~5 times more permeable at 70°C and 8 times at 105°C, when compared to permeability at 21°C.

IV. CONCLUSION

We have deposited Au and Pd on shells while they were agitated to obtain uniform reproducible coatings. We have used the XRF technique to accurately measure thicknesses and uniformity of the deposited layers on shells. We have demonstrated that Au shells are highly reflective, but rather impermeable, while Pd coated shells are substantially more permeable but less reflective. Au coating permeation experiments were performed at higher temperature in an effort to increase the Au coating's permeability. Seeking to combine the permeability of Pd coatings and reflectivity of Au coatings, uniform AuPd alloy coated shells were made. AuPd coatings demonstrated higher permeability compared to Au, and higher reflectivity compared to Pd.

Over the long term we need to measure reflectivity of actual shells instead of flats, measure reflectivity in cryogenic conditions, examine coating actual foam shells to be used in IFE, and adjust temperature at which shells are permeation filled in order to find optimal permeation conditions.

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