

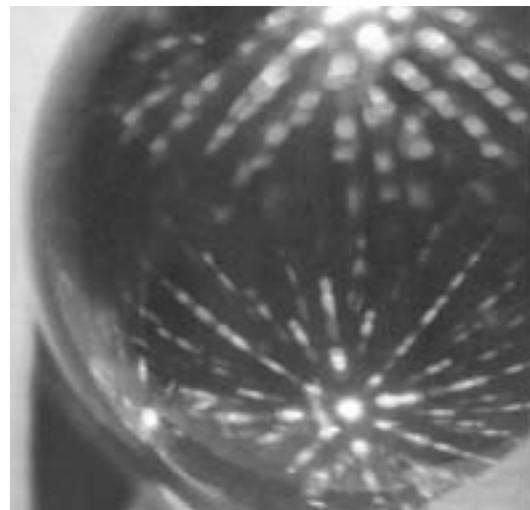
Palladium Coatings of Targets for Inertial Fusion Energy

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Whether arguing about geocentric orbits or creating stories of rebellious gods cast into the heavens for their deeds, humans have needed to understand and rationalize the heavens. The ancient Mayans revered the stars and, in some sense, conquered them by forming incredibly accurate models of the heavens. In the field of nuclear fusion today, scientists have a new dream for star conquest: to harness the energy of the stars to fuel our human world. General Atomics is working to produce a viable fusion energy plant within the next 20 to 40 years. The specific goal may be simply stated. Work in the form of electromagnetic energy is put in to get energy out, and the net must be maximized. The energy utilized is what binds the very nuclei.

Fusion promises to offer a clean (minimal radioactive by-products), inexpensive, and abundant (obtained from water) source of energy for the future. Inertial Fusion Energy (IFE),^{1,2,3} which involves compression of a small (~ 2 to 4 mm in diameter) deuterium-tritium-filled (DT-filled) shell, the “target,” by a driver such as a high-power laser, is an alternative to the more conventional magnetic fusion scheme. In order for this to be profitable, the process must occur six times per second and cost less than 30 cents. The shells also need to have shell walls on the order of tens of microns and uniformity within 10 percent. These are very challenging constraints.

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 preheat direct drive target” (Fig. 1). The shell can be thought of as an inner portion of fuel (payload) surrounded by an ablator with hi-Z rich exterior. Laser beams are used to vaporize the ablator and reaction forces from the ejected gases compress its contents. But as the laser vaporizes the hi-Z coating, the hot



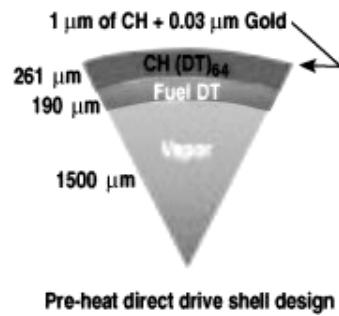


Figure 1

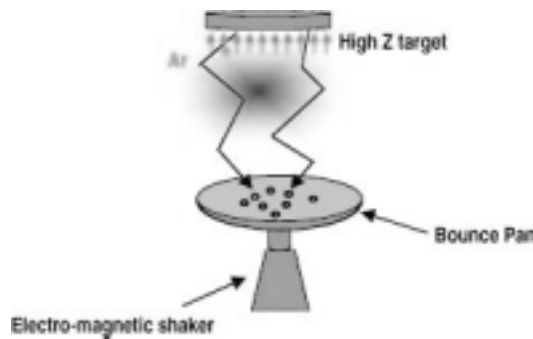


Figure 2

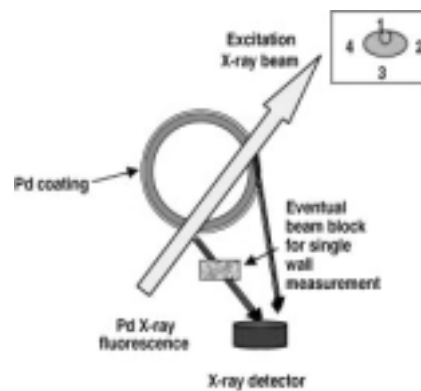


Figure 3

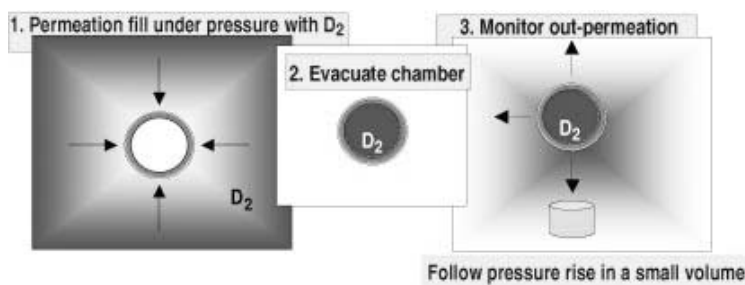


Figure 4

plasma emits a short burst of x-rays that penetrates beneath the vaporization front and, by heating the ablator, stabilizes it against instabilities. But the target first must get to the center of the hot reaction chamber without the fuel layer melting. By concentrating the hi-Z material into a surface coating and choosing a reflective metal for the element, this component could also protect the frozen fuel from exposure to blackbody radiation of the chamber. The chamber must be hot in order to maximize thermal efficiency of the power plant, as governed by Carnot cycles. High reflectivity of such a coating is required in order to tolerate the blackbody radiation from such a chamber. Therefore, coatings with high reflectivities are desired. Secondly, the coating of the shell must enable a quick fill time. Because of the hazards of tritium, its degradation into He3 creating bubbles, and the practical functioning of a power plant needing to shoot six shells per second, the fill time must be short and the tritium inventory kept low. Knowing that the fill time is directly proportional to shell permeability,⁵ ideally the shell must be quite permeable.

Previous work involved the use of gold as the overcoating for its excellent reflectivity in the optical wavelength region of interest (~ 0.5 to $25 \mu\text{m}$). Au coatings, however, had proven to be not permeable enough for a practical power plant. We chose to investigate Pd because of its known high permeability to hydrogen and its isotopes.^{6,7} This particular metal had also proved promising in the computer simulations. Pd reflectivity is known to be lower than Au, but there were significant discrepancies in previous works on Pd reflectivity.⁸ Our work addressed that question as well.

The work reported in this paper describes deposition and characterization of palladium as the hi-Z material. We have deposited Pd on shells while they were agitated to obtain uniform reproducible coatings. We have 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, the previous hi-Z material used. While Pd coatings on flats deformed and cracked, Pd coatings on shells remained stable upon extended and repeated exposure to the surrogate fusion fuel, D2. The main disadvantage of Pd coatings compared to Au is their lower reflectivity, which leads to a lower working temperature of the proposed fusion reactor.

II. Experimental Techniques

A. Deposition Pd coatings were sputtered onto targets.^{9,10} 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 PAMS (poly-alpha methyl styrene) shells. 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 gunpower, gun-substrate distance, deposition pressure, and shell agitation. The pressure and gun-substrate distance are 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, resulting in less dense and reflective coatings. 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 PAMS shells cannot be brought too close or they will melt. We used the deposition condition on flats and shells for consistency.

B. Thickness Measurement by X-Ray Fluorescence (XRF) (Fig. 2) Since the thickness of Pd needs to be a narrow range (~ 300 to 1000 Å), it is crucial to measure the coating thickness accurately. In addition, we needed to ascertain any Pd thickness nonuniformity around the shells. Coatings are traditionally measured using profilometry, which simply traces the profile of the film where a portion has been scratched off. It is practically a step measurement. Profilometry, while possible on flats, is impractical to use on shells. A witness plate can be used for each coating run alongside the shells.¹¹ 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; because these shells are bouncing randomly, the entire shell surface is coated. But the scattering of sputtered material on the way to the shells can alter this factor from the theoretical value of four. Furthermore, it is difficult to position the witness plate correctly, because it can be placed only at the edge of the pan. When the witness plate is placed at the same height as the shells, it is inaccurate due to shadowing of the edge of the pan.

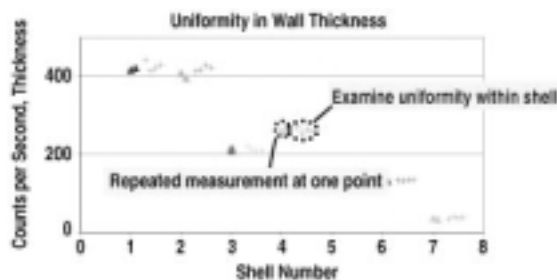


Figure 5a

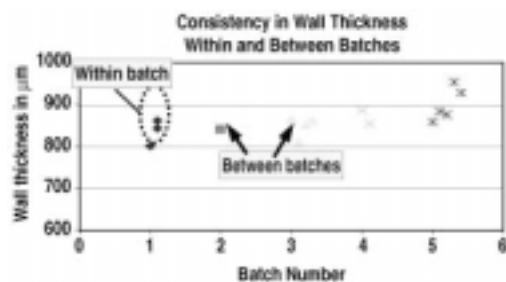


Figure 5b

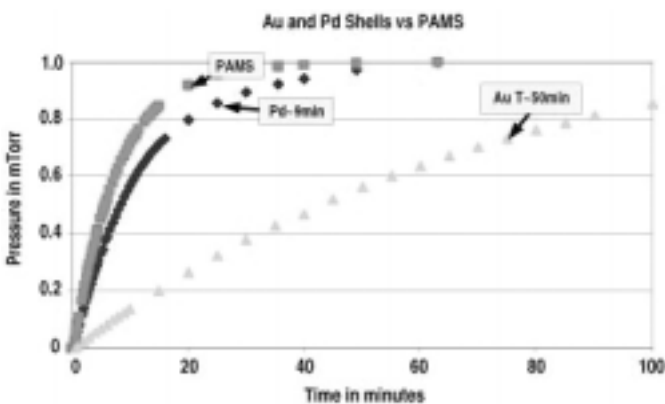


Figure 6

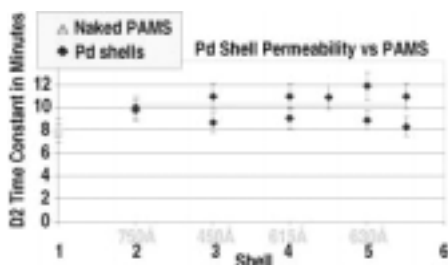


Figure 7

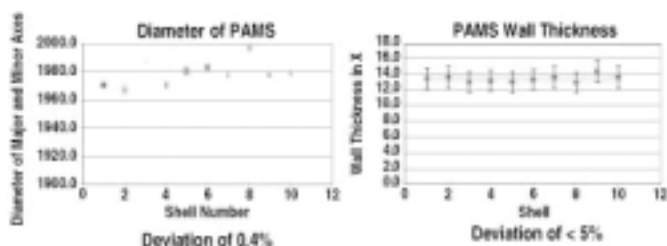


Figure 8

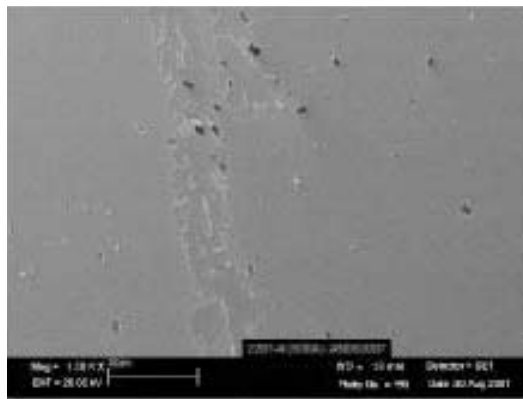
Further Reading

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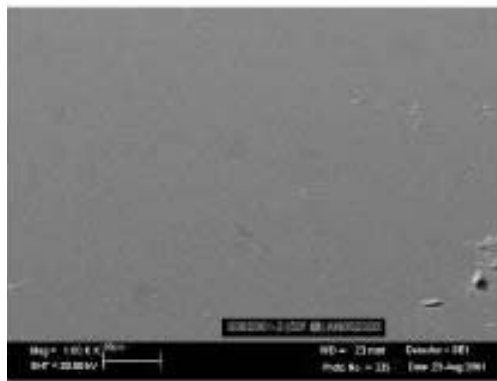
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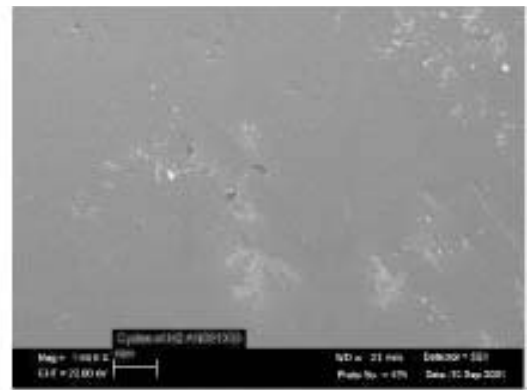
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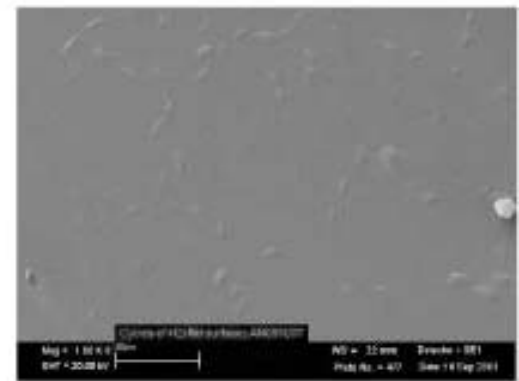
1. Shell before



1. Flat before



2. Shell after D2 exposure



2. Flat after D2 exposure, residual defects.

Figure 9

On the other hand, if it is placed such that there is no shadowing, it is significantly higher than the shells. In any case, the witness plate is not in the same condition as the shells. In addition, no thickness uniformity information on the shells can be obtained when using a witness plate.

XRF provides a relatively simple, non-destructive method to determine the amount of metal deposited. In this method the sample is hit with an x-ray beam. As a result, the metal is excited and emits x-rays characteristic of the elements present in the sample. A detector captures a portion of these x-rays and the resulting peaks correspond to the amount of element present in the sample.¹² In measuring shells, the beam hits two spots on the shells (Fig. 3). 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 the 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 also traverse the PAMS, but we found that there was no attenuation from the PAMS.¹³

C. Permeability Measurements (Fig. 4) 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 almost completely filled with gas, the chamber was evacuated. Then the pressure rise in the chamber resulting from the outgassing of the shell was monitored. This rise in pressure was then fitted to an exponential function to determine a permeation half-life. Background outgassing from gas absorbed in the chamber walls was taken into account when appropriate.^{14,15}

III. Results and Discussion

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 thus is frequently used as a catalyst and in purifying hydrogen. However, Pd films are also known to greatly

deform and crack under exposure to hydrogen due to a large expansion of their crystal structure.^{16, 17, 18, 19, 20}

We successfully coated PAMS 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. 5a). We proceeded to check the shell-against-shell uniformity within a given batch (Fig. 5b). These measurements showed that the shells within a batch were uniform within the measurement error of about 10 percent. We then wanted to verify that shells of different batches were consistent. In analyzing shells of different batches we also found consistency of about 10 percent. So the shells produced were uniform and reproducible.

We used palladium for its permeability to hydrogen and its isotopes, which our experiments confirmed. Due to radioactivity issues with DT, we used deuterium (D₂) gas as a surrogate for the permeability measurements. Figure 6 shows that outgassing data for deuterium through a bare PAMS shell and an ~ 1100Å Pd-coated PAMS shell are very similar. In general, Pd-coated shells have a permeation time constant of about 10 ± 1 minute, only slightly less permeable than the bare PAMS (~ 8 ± 1 min). Therefore, Pd coatings are indeed very permeable when compared to gold coatings, which had time constants of ~ 1 hour for ~ 250Å coatings, as previously measured. Interestingly, we found that the permeability of Pd shells did not depend on the coating thickness. Shells of considerably different thickness exhibited the same time constant (Fig. 7). This implies that there may be a limiting step such as disassociation of the D₂ into Pd and then the gas would quickly pass through to the center. The variation of permeability between shells was within measurement errors, but any variation of the mandrels underneath the Pd would contribute to our measured variations in permeability of palladium shells. In order to determine how much of the variation was caused by the mandrels, we measured the variation in diameter and wall thickness of the mandrels. These two factors directly determine the time constant. From this analysis we found that variation in diameter contributed a 0.5 percent error, while variation in wall thickness contributed a 5

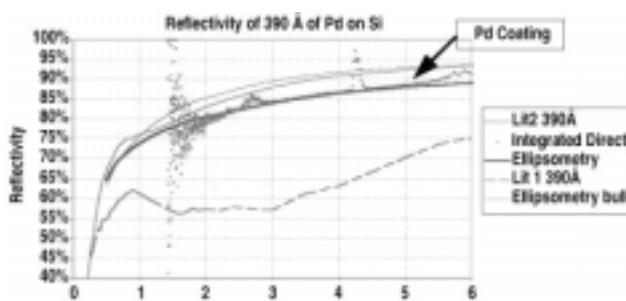


Figure 10

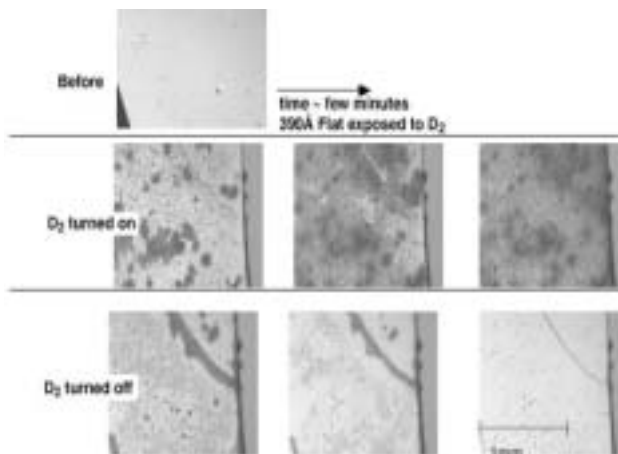


Figure 11

percent error (Fig. 8). These errors, therefore, contribute to the 10 percent error we saw in Pd-coated shells, but it seems that some of the variation is due to the palladium coating itself.

In order to test for deformation, we coated a glass plate with palladium and exposed it to D₂ for a period of several minutes. Immediately, the coating exhibited gross deformities that grew worse over time. When the D₂ exposure stopped, the coating began to lose its deformities, but the coating on the plate never returned to its original state (Fig. 9). In performing the same experiment with palladium-coated shells, we found no visual deformations. SEM examination of the shells and flats confirmed these results. While the shells displayed the same amount of nonuniformity before and after exposure, the flats showed residual defects from D₂ exposure (Fig. 10). Furthermore, we saw no deformation in the shells that were repeatedly exposed to D₂ for longer periods of time in permeation measurements. From these results, we inferred that filling with DT should be rapid and cause no damage to the Pd coating layer.

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

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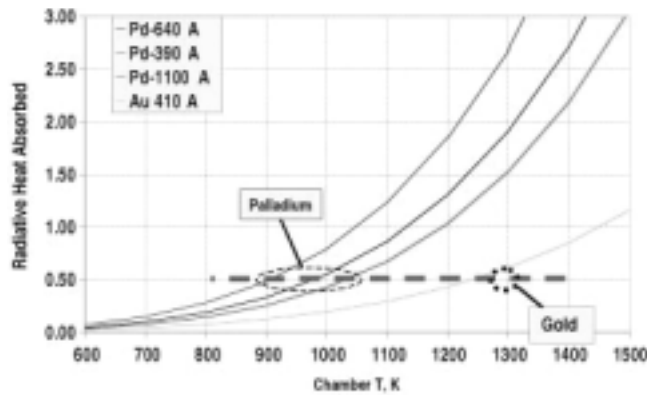


Figure 12

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for palladium, they were still below that of gold (Fig. 11). An important feature to notice is the large spread in the literature values. This demonstrates the effects of different deposition conditions and working methods on reflectivity measurements.²¹ The measurements were done on flats using two independent methods, ellipsometry and integrated direct measurement. In ellipsometry, the material's optical constants, n and k values, are found and can then be used to determine reflectivity at arbitrary angles of incidence using the Fresnel reflection equations.^{22, 23} The second 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 thus they are adequate for comparisons only. Ellipsometry results provide reflectivities at all angles, which are needed for calculation of the heat load on the target because it experiences the omnidirectional radiant heat as it traverses the hot reactor chamber before it gets shot by the driver beams.

We convolved the data with a blackbody spectrum 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,^{24, 25} we found the maximum chamber temperature for which shells coated by various thicknesses of Pd would survive the

chamber heat (Fig. 12). The calculated values for gold-coated ($\sim 300\text{\AA}$) shells are also shown. While the gold shells could survive at a maximum temperature of 1,300K, the optically opaque palladium shells could survive at a maximum of 1,100K. Although this is a disadvantage of the Pd coatings, we hope that by using mixtures of Pd and Au we can form coatings that have high permeabilities, due to Pd presence, and high reflectivities, due to Au presence. In fact, initial results on this work are encouraging and will be reported in a future publication.

IV. Conclusion

We deposited Pd on shells while they were agitated to obtain uniform reproducible coatings; we used the XRF technique to accurately measure thickness and uniformity of the deposited layers on shells; and we demonstrated that these palladium-coated shells are indeed substantially more permeable than gold. While Pd coatings on flats deformed and cracked, Pd coatings on shells remained stable upon extended and repeated exposure to D2. The main disadvantage of Pd coatings compared to gold is their lower reflectivity. We hope that adding gold to the Pd coating will increase reflectivity while maintaining high permeability. Initial results on this work have been encouraging.

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 permeation temperature in order to find optimal permeation conditions.

Acknowledgments:

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