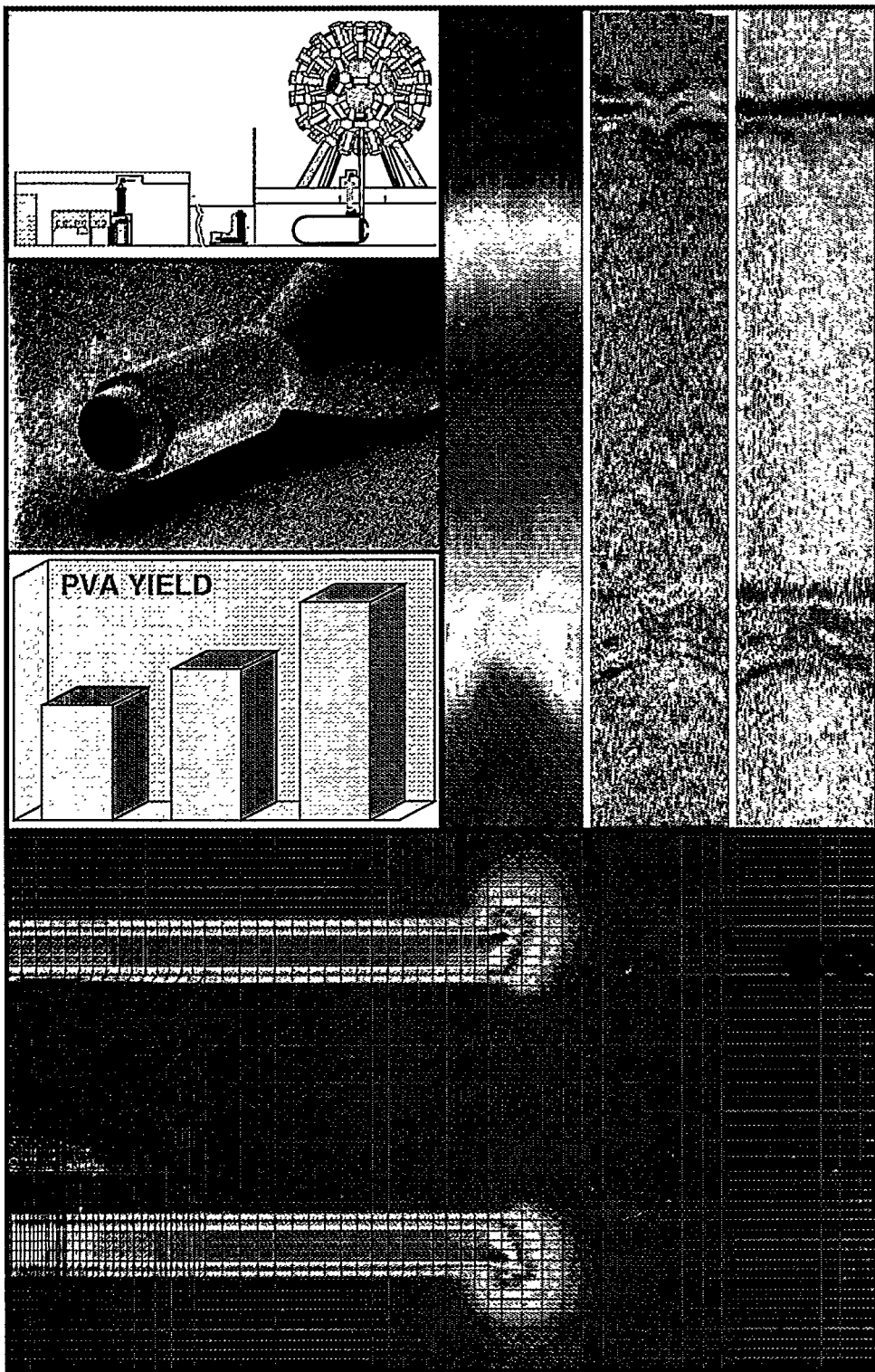


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INERTIAL CONFINEMENT FUSION

ANNUAL REPORT

OCTOBER 1, 1993
THROUGH
SEPTEMBER 30, 1994

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INERTIAL CONFINEMENT FUSION TARGET COMPONENT FABRICATION AND TECHNOLOGY DEVELOPMENT SUPPORT

**ANNUAL REPORT TO THE
U.S. DEPARTMENT OF ENERGY**

OCTOBER 1, 1993 THROUGH SEPTEMBER 30, 1994

by
PROJECT STAFF
Martin Hoppe, editor

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MASTER

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ABSTRACT

On December 30, 1990, the U.S. Department of Energy entered into a contract with General Atomics (GA) to be the Inertial Confinement Fusion (ICF) Target Component Fabrication and Technology Development Support contractor. This report documents the technical activities which took place under this contract during the period of October 1, 1993 through September 30, 1994. During this period, GA was assigned 17 tasks in support of the Inertial Confinement Fusion program and its laboratories. In addition GA was asked to provide direct "Onsite Support" at LLNL, LANL, SNL-A, and UR/LLE.

This year we achieved full production capabilities for the micromachining, dimensional characterization and gold plating of hohlraums. We fabricated and delivered 726 gold-plated mandrels of 27 different types to LLNL and 48 gold-plated mandrels of two different types to LANL.

We achieved full production capabilities in composite capsule production and delivered in excess of 240 composite capsules. In anticipation of increased delivery rates and more technically challenging capsule specifications we continuously work to improve performance and capabilities. We have instituted an inventory system to improve capsule request turn-around time, improved the PVA coating process, developed new doped polymers and coatings, and have added additional and/or improved existing characterization capabilities.

We were also directed to dismantle, remove, and disposition all equipment at the previous contractor (KMSF) that had radioactive contamination levels low enough that they could be exposed to the general public without radiological constraints. GA was also directed to receive and store the tritium fill equipment.

We assisted LANL in the development of techniques for characterization of opaque targets. We developed deuterated and UV-opaque polymers for use by the University of Rochester's Laboratory for Laser Energetics (UR/LLE) and devised a triple-orifice droplet generator to demonstrate the controlled-mass nature of the microencapsulation process.

The ICF program is anticipating experiments at NIF and the Omega Upgrade. Both facilities will require capsules containing layered D₂ or D-T fuel. We are part of the National Cryogenic Target Program to create and demonstrate viable ways to generate and characterize cryogenic layers. We investigate thermal gradient layering for deuterium fuel and plasma-assisted layering for D-T fuel. We pursued optical techniques for measuring the surface finish of fuel layers, and spatially resolved NMR imaging to determine the extent of D-T fractionation.

We continued engineering and assembly of equipment for a cryogenic target handling system for UR/LLE that will fill, transport, layer, and characterize targets filled with cryogenic deuterium (DD) or deuterium-tritium (D-T) fuel, and insert these cryogenic targets into the OMEGA Upgrade target chamber for laser implosion experiments.

This report summarizes and documents the technical progress made on these tasks.

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SECTION 1

TARGET FABRICATION PROGRAM OVERVIEW

1. TARGET FABRICATION PROGRAM OVERVIEW

On December 30, 1990, the U.S. Department of Energy entered into a contract with General Atomics (GA) to be the Inertial Confinement Fusion (ICF) Target Component Fabrication and Technology Development Support contractor. This report documents the technical activities of the period October 1, 1993 through September 30, 1994. GA was assisted by W.J. Schafer Associates (WJSA) and Soane Technologies, Inc. (STI) and we have carried out the ICF Target Fabrication tasks as a fully integrated team effort.

1.1. INTRODUCTION

In FY94, the GA/WJSA/STI team maintained previous production capabilities in all areas of ICF target fabrication. In addition, in several areas of target fabrication we have developed and/or added new capabilities.

1.2. FY94 TASKS SUMMARY

During FY94, the GA/WJSA/STI team was assigned 17 formal tasks, as shown on Table 1-1. These tasks are described briefly here. Additional technical detail on selected topics is given in Section 2 through 6 of this report.

1.2.1. GA02 EQUIPMENT REMOVAL FROM KMSF

The previous ICF Target Component Fabrication and Technology Development Support contractor, KMS Fusion, accumulated considerable Department of Energy (DOE-) owned equipment over the course of its contract. DOE planned to deliver this equipment to General Atomics during the transition year 1991. Unfortunately, disputes between KMS Fusion and DOE kept the bulk of the equipment under the control of KMSF until January 1994.

Table 1-1
FY94 Target Fabrication Tasks

Task #	Task Title/Description	Total \$K	Task Leader
GA02	Equipment Removal from KMSF	370	Miller
LL01	On-Site Support at LLNL	530	Miller
LL02	Micromachined Target Components	1,036	Shillito, Kaae
LL03	Characterization Development	133	Sater
LL04	Composite Polymer Shells	760	Miller
LA01	On-Site Support for LANL (@LLNL)	137	Miller
LA02	Composite Polymer Shells	760	Miller
LA03	Polymer Shell Prod. by Microencaps.	400	Nelson
LA04	Glass Shell Deliveries	50	Miller
NR01	NRL Target Development and Delivery	980	Hendricks
SL01	Fab. of Foils, Filters and Polymers	1,036	Kaae
UR01	LLE Materials Development	96	McQuillan
CR/LL1	Cryogenic Layering Development	885	Bittner
CR/LA1	Beta Layering Support at LANL	200	Simpson
CR/LA2	Cryogenic Layering Technology	178	Alexander
CR/UR1	OMEGA-U Target System Engineering	1,583	Fagaly
CR/UR2	Cryo. Characterization and Foam Dev.	470	Stephens
Total:		\$9,604	

DOE directed GA to dismantle, remove, and disposition all equipment that had radioactive contamination levels low enough that they could be exposed to the general public without radiological constraints. GA was also directed to receive and store the tritium fill equipment. These activities needed to be coordinated with the DOE/SF property manager and the DOE-appointed site manager.

In January, GA visited the former KMSF site, photographed the equipment in each room and identified useful equipment. We prepared plans for removing the equipment and coordinated them with the DOE/SF property manager, with Battelle Pacific

Northwest Laboratories (DOE's initial site manager), and later with Lawrence Livermore National Laboratory (LLNL) (DOE's final site manager). In spring, we dismantled equipment including the drop tower furnaces for making glass and polymer shells, coaters for depositing inorganic and polymer layers, analytical equipment, and other miscellaneous items. In early June, we packed and shipped most of the equipment required by the ICF program to sites in California and New Mexico. The remaining few items were shipped to GA in late summer. Many pieces of equipment have already been installed, made operational, and are being used in support of the program.

1.2.2. LL01 ONSITE SUPPORT FOR LLNL

Our onsite team at LLNL emphasized activities in micromachining of target components, assembling target component in complete targets, and characterizing target components to assure their quality for use in NOVA ICF experiments. Craig Rivers micromachined many types of target components. John Ruppe and Steve Grant, who started in February, assembled and characterized over 350 targets. Andrea Denton-Paul characterized capsules and target components using several techniques.

In addition to these target manufacturing activities, two senior scientists, Fred Lim and Chuck Hendricks, were consulted by LLNL on activities relating to capsule R&D.

1.2.3. LL02 MICROMACHINED TARGET COMPONENTS

We achieved full production capability for the micromachining, dimensional characterization and gold plating of hohlraums this year. In all, we delivered 774 gold plated mandrels of 29 different types to LLNL and Los Alamos National Laboratory (LANL). We also added to our micromachining capabilities with the purchase of a milling attachment for the Precitech 2000 lathe. This attachment greatly increases the speed with which flats can be machined on hohlraums. In addition General Atomics purchased a second Precitech Optimum 2000 lathe with company funds. This lathe is also available for ICF micromachining tasks, which further adds to our micromachining capabilities.

Micromachined components other than hohlraums include witness plates, Rayleigh-Taylor samples, composite (metals and plastics) mix packages, and miscellaneous one-of-a-kind physics packages. The first development toward witness plate validation involved preparation of aluminum witness plates. However, before validation was

achieved on Al witness plates the priorities were changed and we were asked to produce plastic witness plates from polycarbonate or polystyrene material. Methods investigated for the preparation of plastic witness plates include thermally-pressed polycarbonate, cast polystyrene solutions in aluminum molds, direct machining into a cast or thermally-pressed polymer, and direct machining into a thin polymer film. While the results have yet to fully meet the desired specifications, at least partially due to continuing mechanical problems with the Rocky Flats #3 lathe, the results were encouraging. Efforts in producing plastic and metal witness plates will continue into next year. The machining programs required for the Rayleigh-Taylor samples were also prepared and validated.

1.2.4. LL03 CHARACTERIZATION DEVELOPMENT

This task was comprised of three characterization development subtasks, each of which are summarized below.

Photo-Nuclear Reactions Measurement Methods. A study was undertaken to evaluate the potential of gamma-neutron reactions to measure the deuterium content of filled inertial confinement fusion capsules. Because various hydrogen isotopes (hydrogen, deuterium, and tritium) have different diffusion rates, the relative isotope concentrations in a capsule prior to reaching equilibrium pressure can vary from the relative concentrations used to fill. A non-destructive method of analyzing the fill concentrations in a capsule could also be used to detect fuel loss due to capsule leaks.

We found that gamma-neutron reactions using a gamma source with energies above 2.23 MeV and source strength on the order of a Curie could be used to measure the deuterium content of a filled capsule. This measurement would be non-destructive. However, the cost of setting up and operating an experimental area using a gamma source with the required strength is prohibitive. This led to a recommendation not to pursue an experiment at the present time. This measurement technique will only be practical if affordable access to an existing high-energy gamma source can be arranged.

Computer Assisted Shell Selector (CASS). Polystyrene shells are used as the mandrel in current ICF composite plastic capsule designs. About 2% of the polystyrene mandrels make it through the various coating steps to become finished capsules. The low yield and large number of finished shells needed require a technician to characterize many mandrels for any given delivery. The characterization can be repetitious and time

consuming. The Computer Assisted Shell Selector (CASS) was proposed by Joe Mauger, at LLNL, as an instrument to help automate initial characterization and selection of polystyrene shells. A prototype CASS was constructed at LLNL. During FY94 we completed and tested this prototype.

The objectives of the CASS system are to:

1. Characterize the diameter and eccentricity of mandrels.
2. Measure wall thickness of the shells.
3. Remove the accepted shells for continuation on the successive steps in the fabrication process.

We ran tests to determine precision in the diameter measurement. Over several runs the system consistently identified shells. The shells were also removed from the mirror surface and manually measured by Charlotte King. The average difference in diameter measurement was $3.1\text{ }\mu\text{m}$, less than 1%. We also performed a series of runs to time the diameter and circularity measurements. We found it would take 5 hours to scan a 4x4 inch surface containing 2000 shells assuming half of them were good. Wall thickness measurement takes up to 3 minutes per shell. Charlotte King's measurements of a sample of shells differed from the CASS measurements by a range of $0.02\text{ }\mu\text{m}$ to $0.54\text{ }\mu\text{m}$ with an average of $0.27\text{ }\mu\text{m}$, about 10%.

The possibility of using CASS as a production tool at General Atomics was investigated. General Atomics characterizes polystyrene drop-tower shells by sieving. CASS can not keep up with the selection speed possible by sieving and should not be pursued any further if this were its only possible function.

Micro-encapsulated shells and glass shells are not as uniform in wall thickness as drop-tower polymer shells. The CASS wall thickness measurement capability may be of use in this case. CASS's capability to measure parameters of a large number of shells may also be useful in carrying out screening studies of shell production processes.

Micro-Calorimetric Determination of ICF Capsule Fills. There is a need in the ICF program for a means to non-destructively determine the amount of fuel contained in a laser fusion target. Several techniques exist for determining this parameter but they rely on the capsule to be transparent. We have developed a unique means to non-destructively

analyze opaque targets as well as transparent ones. Our technique uses a calorimetric method to measure the fuel content. We cool a single, filled capsule and use the change in heat capacity of hydrogen as it condenses to accurately determine the fill pressure. We have successfully measured the fill of an 800 μm diameter glass capsule filled with 50 atmospheres of D_2 . By scaling based on our signal to noise, we have estimated an upper limit for our sensitivity. We estimate it is possible to detect a measured change of 2×10^{-6} Joules/Kelvin in heat capacity. This allows one to measure the fill of an 800 micron diameter capsule containing 1.3 atmospheres of D_2 . If we rescale these results to a 400 micron diameter capsule, we calculate 10 atmospheres to be the detection limit of the current apparatus. This technique should be developed into a production tool for measuring capsule fills. We also made specific recommendations for improvement of our instrument.

1.2.5. LL04 COMPOSITE POLYMER SHELLS FOR LLNL

The goal of this task was to manufacture, characterize, and deliver low-Z fuel capsules required by LLNL and to develop improvements in the manufacturing process. GA made six capsule deliveries with specifications shown in Table 1-2. In nearly every case we delivered the number of shells requested or even more (Fig. 1-1). The quality of the capsules was excellent as determined by comparing the actual tolerances of the capsules to the specified tolerances as shown by Table 1-3.

An important part of this task was developing new capabilities and improving current capabilities so that GA will respond even better to Laboratory requests. Among these were developing techniques for responding rapidly to urgent Laboratory requests, manufacturing capsules doped with titanium and/or germanium, and new characterization capabilities for measuring layer thicknesses and the elemental composition of key constituents in the capsule.

Rapid Response to Urgent Laboratory Requests. Frequently the nature of the experimental ICF program provides little lead time from when the specifications for a capsule are provided and the date the capsule must be delivered. The process we had been using required about two months lead time. A faster response was needed. The goal was to deliver complete, fully characterized capsules within 21 days of an urgent request. We achieved this in two ways, by improving our processes and by going to an inventory system.

Table 1-2
LLNL Capsule Delivery Specifications

Item	Shell Type	Mandrel ID (μm)	Poly-styrene (PS) Wall (μm)	Polyvinyl Alcohol (PVA) Wall (μm)	Fill (atm)	Total Wall Thickness (Mandrel + PVA + CH)
LLNL-P03	PS with 1 atom % Cl	420-460	2-4	2-6	0.05 Ar	$43 \pm 2 \mu\text{m}$ CH
LLNL-P04	PS with 0.3 atom % Cl	420-460	2-4	2-6	0.05 Ar	$43 \pm 2 \mu\text{m}$ CH
LLNL-P05	PS with 1 atom % Cl	420-460	2-4	2-6	0.025 Ar	$43 \pm 2 \mu\text{m}$ CH
LLNL-P06	PS	420-460	2-4	2-6	0.12 Ar	$55 \pm 3 \mu\text{m}$ CH
P-16	PS	420-460	2-4	2-6	0.12 Ar	$55 \pm 3 \mu\text{m}$ CH
P-22	PS	420-460	2-4	2-6	0.05 Ar	$58 \pm 3 \mu\text{m}$ CH

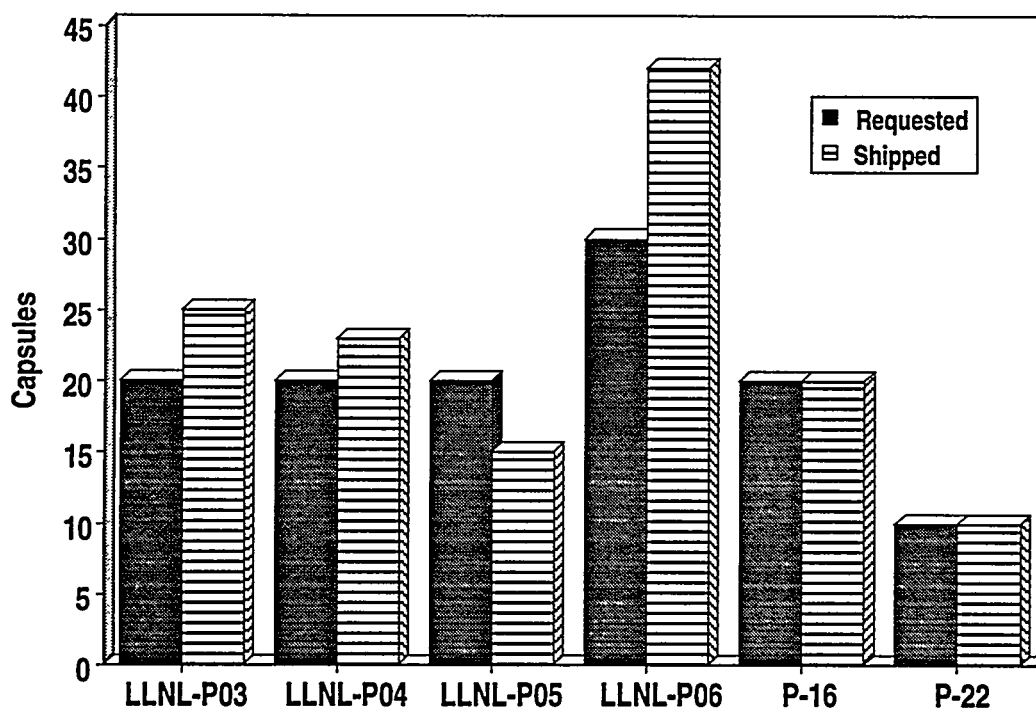


Fig. 1-1. FY94 LLNL capsule orders and deliveries.

**Table 1-3
Capsule Tolerances**

	Specified Tolerance	Typical "Worst in Batch" Tolerance
Inner Diameter	$\pm 20 \mu\text{m}$	$\pm 10 \mu\text{m}$
Total Wall Thickness	$\pm 5 \mu\text{m}$	$\pm 3 \mu\text{m}$
$\Delta w = w_{\text{max}} - w_{\text{min}}$	$\leq 5 \mu\text{m}$	$\leq 3 \mu\text{m}$
<u>DOMES (width)</u>		
> 40 μm	0 ea	0 ea
30–40 μm	2 ea	0 ea
20–30 μm	5 ea	1 ea
10–20 μm	10 ea	3 ea
5–10 μm	20 ea	6 ea

Our least efficient process was coating shells with PVA. The yield was poor, less than 2% of shells injected, and some runs produced no yield at all. Initially, we identified the symptoms of poor yield and then tried to discover and correct the sources. Figure 1-2 shows the relative symptoms. "Strings" of shells stuck together by PVA were frequently a major problem. We found that the frequency of strings dramatically decreased when we did not fill the shells with argon before coating with PVA. Debris was also a major problem. We decreased debris by going to a new fill/injection device and by changing our shell processing laboratory into a clean room. These changes caused our yield to nearly double from 1.9% to 3.6%. Details are provided in Section 2.2.

Filling shells with argon before coating with PVA was not working well. Not only were strings a major problem, but argon pressure varied greatly from capsule to capsule. We discovered that argon could be permeated into capsules as the final manufacturing step, i.e., after both the PVA and glow discharge polymerization (GDP) coating steps. Not only did this increase our overall yield, but it also eliminated the variation in argon pressure between capsules. Details are provided in Section 2.4.

Although our GDP coating operation is reliable, it is time consuming, frequently requiring seven to ten days to coat a typical batch. When several orders must be

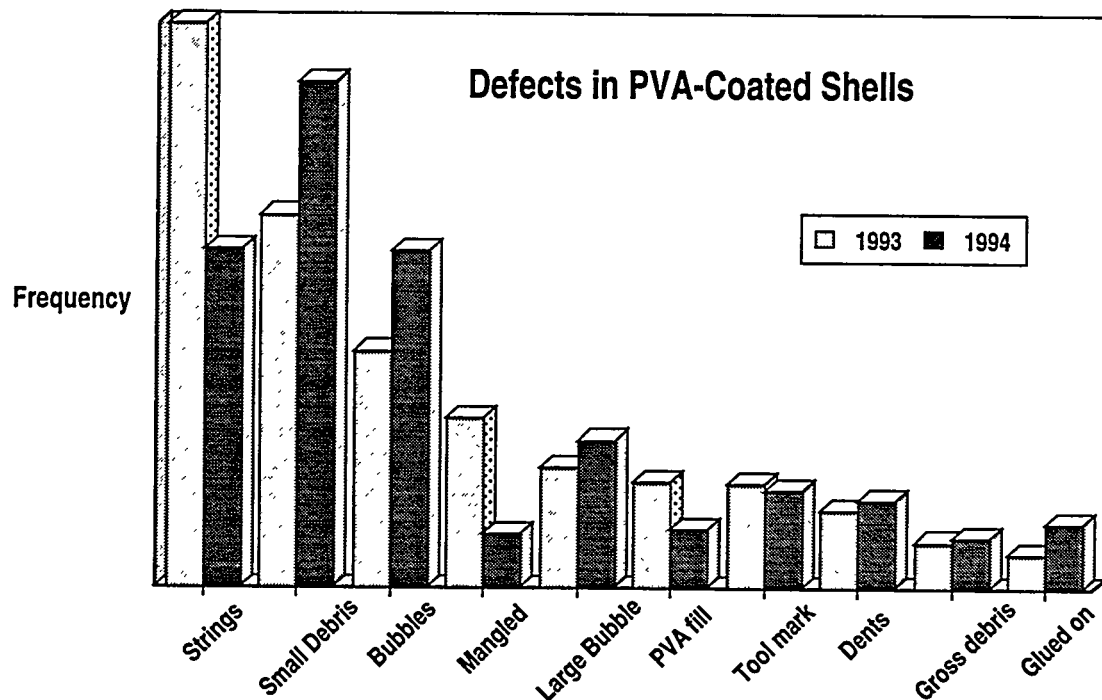


Fig. 1-2. Improvements to defects in PVA-coated shells.

delivered promptly, the GDP process can cause delay. We addressed this by assembling another GDP coater so that two batches can be coated simultaneously. Details on the GDP process are provided in Section 2.2.

We instituted an inventory system to enable us to respond rapidly to Laboratory requests. As can be seen from Table 1-2, capsule orders generally have the same polymer shell and PVA layer thickness specification but differ by the argon content and the total wall thickness. We created an inventory of bare polymer shells, PVA-coated shells and PVA coated shells with a few μm 's of GDP. When faced with an urgent Laboratory request, we pull PVA-coated shells (or the thin GDP coated shells) from inventory and coat them with the required amount of GDP. Thus the slow step with poor yield, coating shells with PVA, does not slow our response.

In early FY95 we tested how well these improvements would decrease our lag time between the date we received the order and the date we shipped the capsules. As shown by Table 1-4, our response time for urgent orders that could be handled by our inventory had decreased from about two months to two weeks.

Table 1-4
Urgent Order Response Time

Item	Requested	Shipped	Ordered	Delivered	Response
P-22 (LLNL)	10	10	9/23/94	9/30/94	7 days
P-23 (LLNL)	10	17	9/23/94	10/7/94	14 days
P-27 (LLNL)	20	22	10/20/94	11/2/94	13 days

Manufacturing Capsules Doped with Titanium and/or Germanium. NOVA experiments require capsules doped with titanium in the mandrel and germanium in the outer CH layer. LLNL had developed processes to make doped capsules, but had not yet perfected them. Titanium-doped mandrels frequently had too little titanium and cracked easily during the assembly of the capsule into the hohlraum. They also found the germanium content in the outer CH layer was difficult to control well enough to get the requested amount of germanium. We transferred the techniques from LLNL and explored changes. Our goal was to demonstrate production capability by October 1.

The LLNL technique for making Ti-doped mandrels was to start with a polymer doped with chemically bound titanium. The University of Alabama (U of A), under contract by LLNL, had examined ways to synthesize the polymer. We contracted with U of A to provide us with polymer. Although we received several batches, none had titanium content as high as the level desired (between 0.06 and 0.2 atom percent). We explored another technique, dissolving a titanium compound into a solution of our usual polystyrene and using this new mixture to make shells. We tried several titanium compounds and found one that worked quite well, titanium (IV) bis(ethylacetoacetatodiisopropoxide). Shells made by this "additive" technique are resistant to cracking and are doped at the desired concentration. However, we are concerned that the titanium might not be homogeneously distributed through the shell wall. Initial analytical tests show no evidence of inhomogeneity, but further testing is required. Details are provided in Section 2.1.

We built, installed, and tested a GDP coater for depositing germanium-doped CH on shells. Although we found that the surface finish became rougher as germanium concentration increased, we were able to deposit good layers and control the germanium

content. By the end of the year we made sample capsules with Ti-doped mandrels and Ge-doped GDP. Details are provided in Section 2.2.

New Characterization Capabilities. Precise and accurate characterization is required on each composite capsule. Characterization includes accurate determination of inner and outer diameters, layer thicknesses, total wall thickness variation (nonuniformity), and surface finish. Characterization also includes chemical analysis for dopants and gases contained within the capsule. At the beginning of the year, we could only measure outer diameter, total wall thickness, total wall thickness variation, and surface finish. This year we added two additional techniques, radiographic image analysis and x-ray fluorescence (XRF), which allow us to perform the rest of the characterization (except for the AFM surface power spectrum). Details are provided in Section 2.3.

1.2.6. LA01 ONSITE SUPPORT FOR LANL AT LLNL

Kett Gifford worked with our onsite team at LLNL, but primarily on LANL targets. He assembled and characterized over 130 targets.

1.2.7. LA02 COMPOSITE POLYMER SHELLS FOR LANL

The goal of this task was to manufacture, characterize, and deliver low-Z fuel capsules required by LANL and to develop improvements in the manufacturing process. GA made six capsule deliveries with specifications shown in Table 1-5. In nearly every case we delivered the number of shells requested or even more (Fig. 1-3). The quality of the capsules was excellent as determined by comparing the actual tolerances of the capsules to the specified tolerances as shown by Table 1-3.

An important part of this task was developing new capabilities and improving existing ones so that GA will respond even better to laboratory requests. The new capabilities and improvements are described in Section 1.2.5.

Two types of capsules were especially challenging, the Symcaps (LANL-P07, LANL-P08, and LANL-P09) and the deuterated mandrel (LANL-P06).

The symcaps presented a special challenge: pairs of shells must be matched by inner diameters (i.d.) to within 4 μm . The bare polystyrene shells were sieved to within about

Table 1-5
LANL Capsule Delivery Specifications

Item	Shell Type	Mandrel ID (μm)	PS Wall (μm)	PVA Wall (μm)	Fill (atm)	Total Wall Thickness (Mandrel + PVA + CH)
LANL-P06	Deuterated PS	420–60	3.5–5.5	2–6	0.1 Ar	$55 \pm 5 \mu\text{m}$ CH
LANL-P07 (Symcap A)	PS	420–460	2–4	2–6	0.8 Ar	$35 \pm 4 \mu\text{m}$ CH
LANL-P08 (Symcap B)	PS	420–460	2–4	2–6	0.8 Ar	$20 \pm 2 \mu\text{m}$ CH
LANL-P09 (Symcap C)	PS	420–460	2–4	2–6	0.8 Ar	$15 \pm 2 \mu\text{m}$ CH
P-17	PS	420–460	2–4	2–6	0.10 Ar	$55 \pm 5 \mu\text{m}$ CH
P-18	PS with 1 atom % Cl	420–460	2–4	2–6	0.10 Ar	$55 \pm 5 \mu\text{m}$ CH
P-19	PS with 1 atom % Cl	150–220	≤ 5	N/A	N/A	N/A

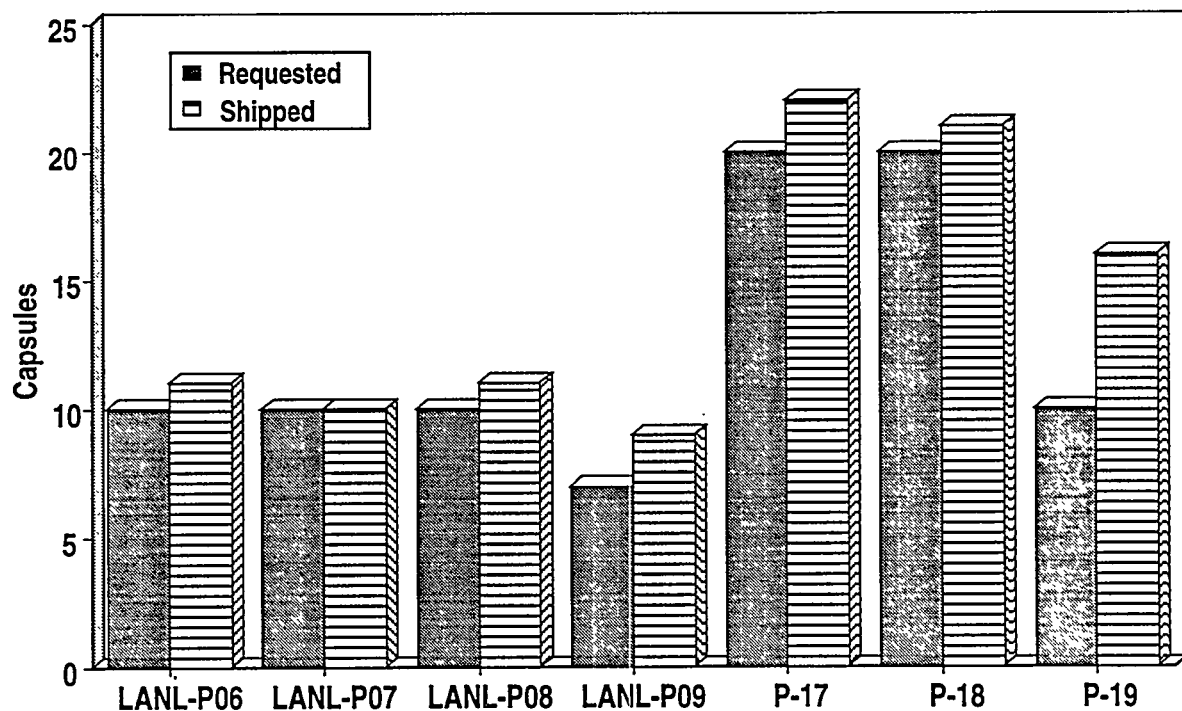


Fig. 1-3. FY94 LANL capsule orders and deliveries.

The specifications for the Symcap (Table 1-6) capsules are:

Table 1-6
Symcap Specifications

	Symcap A	Symcap B	Symcap C
Number of Capsules	10	10	7
Inner Diameter (i.d.)	$440 \pm 20 \mu\text{m}$	$440 \pm 20 \mu\text{m}$	$440 \pm 20 \mu\text{m}$
Mandrel Wall Thickness	$4 \pm 1 \mu\text{m}$	$4 \pm 1 \mu\text{m}$	$4 \pm 1 \mu\text{m}$
Δ I.D. Pairs from different batches (i.e., A, B, and C) must match $ID \leq 4 \mu\text{m}$			
PVA Layer Thickness	$4 \pm 2 \mu\text{m}$	$4 \pm 2 \mu\text{m}$	$4 \pm 2 \mu\text{m}$
Total wall thickness (mandrel + PVA + CH)	$35 \pm 4 \mu\text{m}$	$20 \pm 2 \mu\text{m}$	$15 \pm 2 \mu\text{m}$
Argon	$0.8 \pm 0.15 \text{ atm}$	$0.8 \pm 0.15 \text{ atm}$	$0.8 \pm 0.15 \text{ atm}$
Wall Nonuniformity	$< 2\%$	$< 2\%$	$< 2\%$
Background Surface Finish RMS	$\leq 1000\text{\AA}$	$\leq 1000\text{\AA}$	$\leq 1000\text{\AA}$

$4 \mu\text{m}$ to keep the shell to shell i.d. differences close to the $4 \mu\text{m}$ limit. The problem was measuring the i.d.'s of capsules precisely enough to be certain of a good match. The repeatability of our optical techniques (as measured by the standard deviation) was too large to reliably match shells. However, we were just bringing up the radiographic image analysis technique at the time when characterization of the capsules was needed. The technique worked. We found that the capsules matched even better than required. Although only pairs of shells had to meet the Δ i.d. requirement we found that nearly all the capsules could be matched with any other capsule, shown in Fig. 1-4. Of the 30 capsules delivered, 28 were within $4 \mu\text{m}$ of each other. The overall spread of inner diameters was $5.5 \mu\text{m}$.

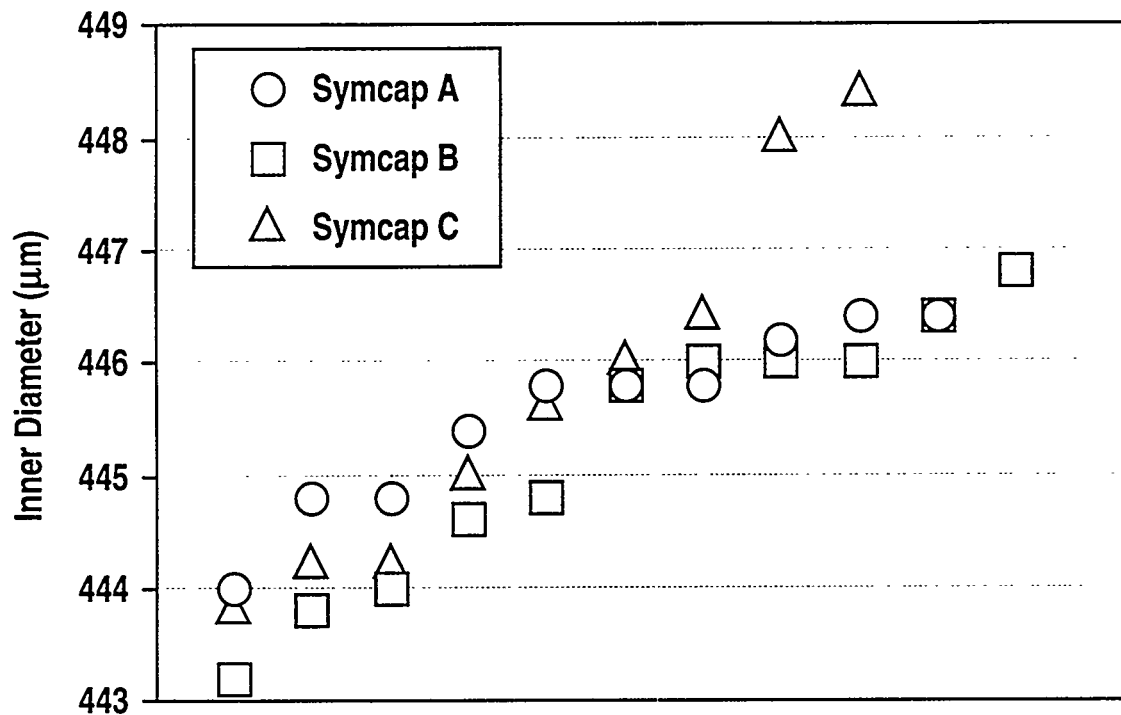
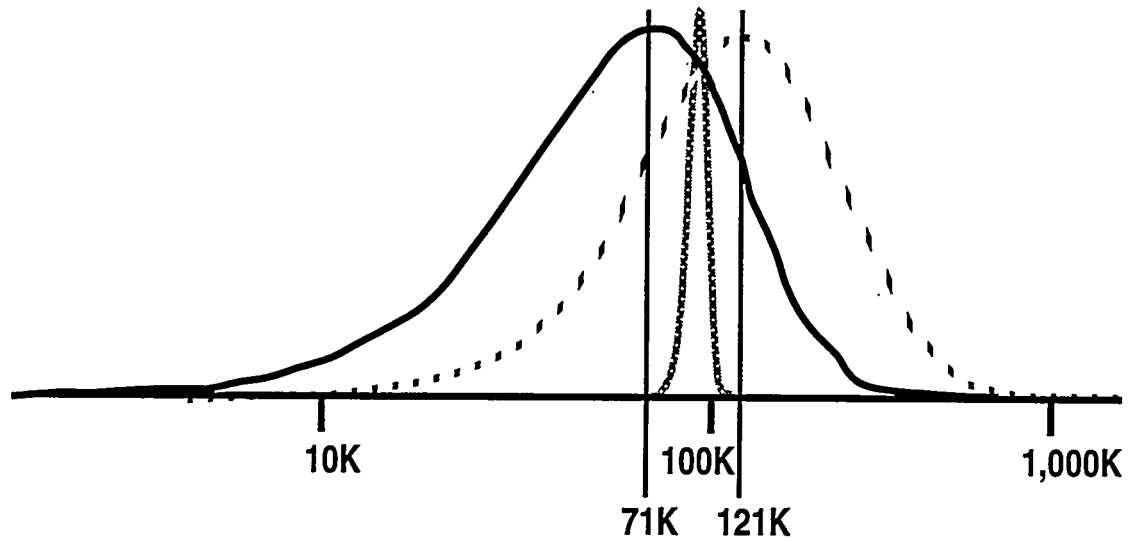


Fig. 1-4. Symcap capsules, showing close matches on diameter.

The LANL-P06 capsules required a mandrel of polystyrene in which all the hydrogen was the isotope deuterium. The deuterated polystyrene required a custom synthesis. We prepared two batches. The batches had a broad molecular weight distribution, shown in Fig. 1-5. We prepared shells by two techniques, drop-tower and microencapsulation. The higher molecular weight batch worked better for microencapsulation; the lower molecular weight batch worked better for the drop-tower. Figure 1-6 shows how the shells compared by interferometry. Our radiographic image analysis shows that capsules with microencapsulation-prepared mandrels have better sphericity and wall uniformity, shown in Fig. 1-7. Unfortunately, the presence of vacuoles in microencapsulation-prepared mandrels makes those capsules less desirable for NOVA experiments.



- Worked best for microencapsulation prepared shells
- Worked best for drop-tower prepared shells
- Usual (normal hydrogen) polymer for drop-tower prepared shells

Fig. 1-5. Molecular weight distribution of deuterated polystyrene LANL-P06 capsules.

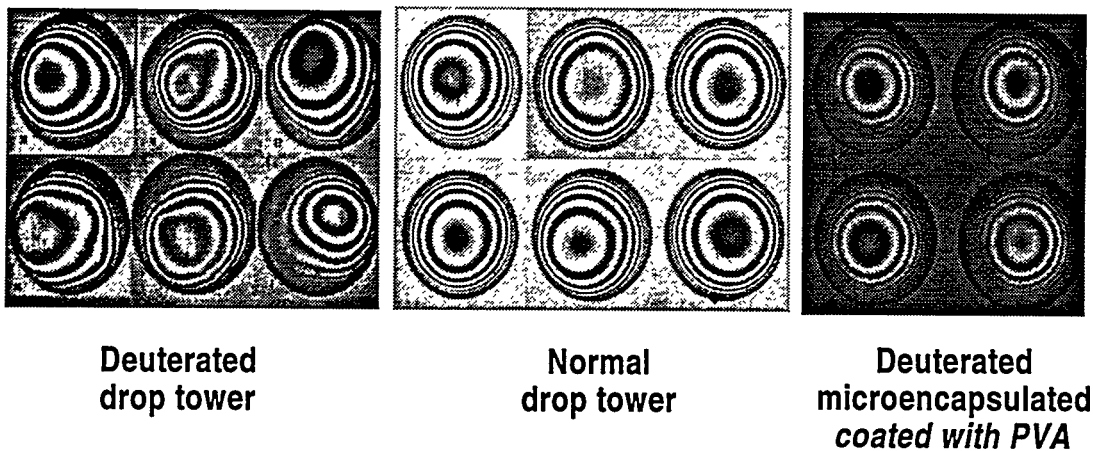


Fig. 1-6. Interference micrographs of drop tower shells made from deuterated polystyrene, drop tower shells made from normal polystyrene and deuterated shells prepared by micro-encapsulation.

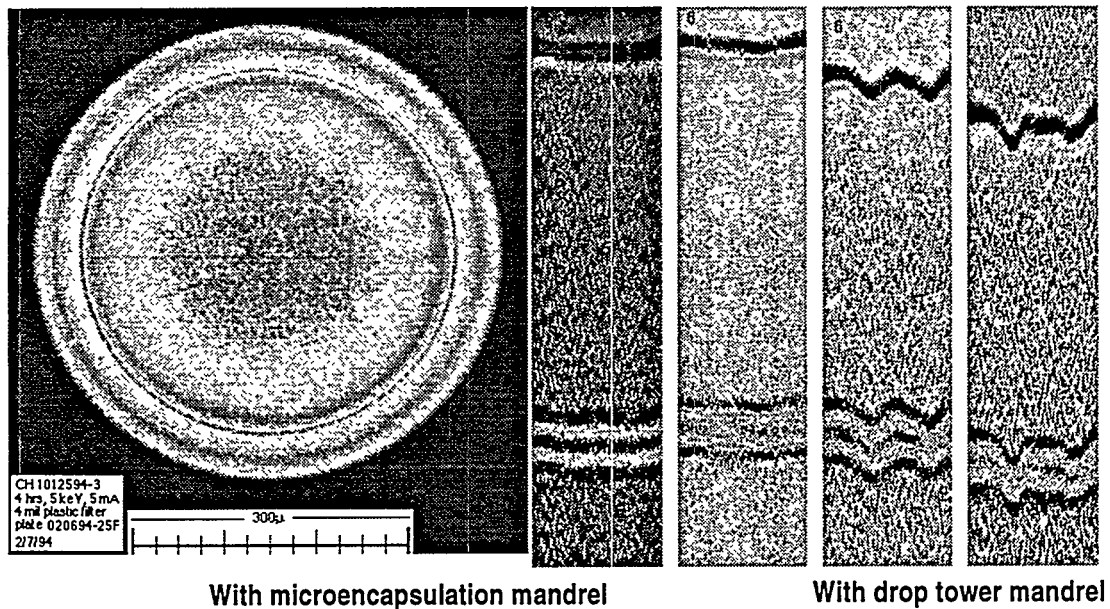


Fig. 1-7. Radiograph from capsules with a deuterated mandrel produced by microencapsulation and prepared by drop tower. The rectangular shaped images are traces of the density variation in a radiographic image. The lines represent interfaces between materials, e.g., between the GDP layer and the PVA layer.

1.2.8. LA03 POLYMER SHELL PRODUCTION BY MICROENCAPSULATION

The overall goal of this task was to develop the ability to produce large polymer shells via controlled-mass microencapsulation. The specific subtasks during the year were:

1. Complete development of the controlled-mass system.
2. Determine optimal operating conditions to produce the best possible millimeter sized shells.
3. Characterize product shells.
4. Complete the mass transfer model, allowing modeling of multi-component solvent systems and the ingression of water into the shell wall.
5. Produce target quality 1.00–1.25 mm o.d. shells with 5–15 μm thick walls.

By attacking these subtasks using a combined experimental and theoretical approach, we were able to achieve nearly all of the goals set at the beginning of the fiscal year for this challenging task. The systematic approach we pursued involved breaking the

problem down into three areas: controlled₇ mass microencapsulation; sphericity and concentricity of the product shells; and vacuole elimination. Finite element mass transfer modeling was particularly important in understanding the role of processing parameters in achieving concentricity. Although experimentally we are usually successful in producing shells with few to no vacuoles, in the future we hope to use this mass transfer model to better understand the processes which govern vacuole formation. While we have met our goals for making low vacuole 1 mm shells, we have not achieved production due to difficulties with debris and drying the fragile shells. After solving debris problems and optimizing the conditions for drying the shells, we expect to be able to produce and deliver large quantities of target quality microencapsulated shells.

1.2.9. LA04 GLASS SHELL DELIVERIES

The goal of this task was to deliver glass shells as required by LANL. Table 1-7 shows the two deliveries that were requested:

Table 1-7
Glass Shell Deliveries

	October Shipment	July Shipment
Outer diameter	$180 \pm 20 \mu\text{m}$	$200 \pm 25 \mu\text{m}$
Wall thickness	$5 \pm 1 \mu\text{m}$	$1.5 \pm 0.5 \mu\text{m}$
Argon pressure	N/A	0.17 to 0.41 atm
Deuterium pressure	N/A	$10 \pm 2.5 \text{ atm}$
Shells requested (delivered)	40 (44)	7 (10)

Of the two, the July shipment was the most challenging since the shells were filled with argon and deuterium. Argon is only slightly permeable through glass, and then only at elevated temperature and with glass that is nearly pure silica. We selected shells with $\geq 99\%$ SiO_2 and permeated them with argon at 600°C and 30 atm for 3 days. Three days is not long enough to get to equilibrium, but was long enough to fill with a few tenth's of an atmosphere. We analyzed for argon by XRF. Shells were then filled with deuterium at 250°C for about 16 hours, conditions sufficient to achieve equilibrium. Deuterium will out-permeate at room temperature. At the temperature of dry ice, the deuterium leak rate is insignificant. We shipped the shells in an aluminum egg-crate cooled with dry ice.

Also under this task we installed and tested the 1700°C drop tower furnace for glass shells obtained from KMSF Fusion. We prepared three batches of glass shells as shown in Fig. 1-8. Although these three batches cover only part of the diameter and wall thickness range achieved by KMSF, we believe we can cover the full KMSF range when needed by the ICF program.

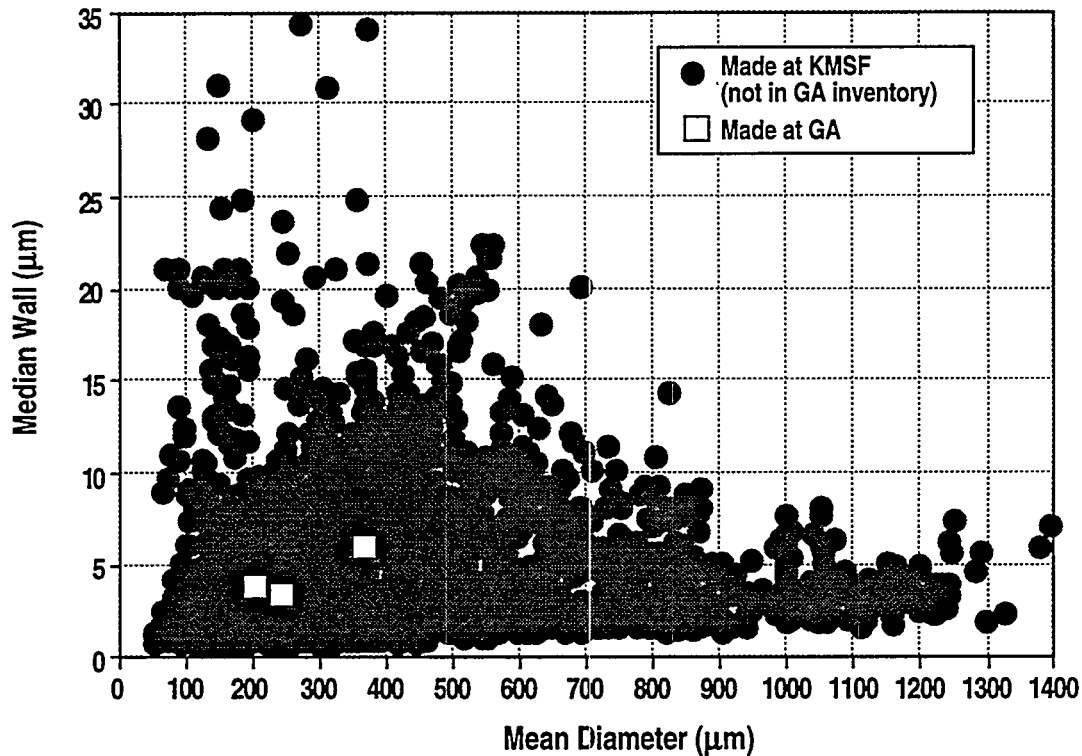


Fig. 1-8. GA tested the 1700°C drop tower furnace obtained from KMSF, making three batches of glass shells. We can make glass shells over the same range as KMSF.

1.2.10. NR01 TARGET DEVELOPMENT FOR NIKE

During the FY94 year our task was to develop the necessary techniques and methods for producing and characterizing NIKE target parts and completed targets and delivering those targets to NRL. The subtasks were (a) to develop fabrication techniques for double CH foils and to deliver prototype mounted double foil targets for NIKE, (b) to develop fabrication techniques for complex CH targets and to deliver prototype complex targets to NIKE, and (c) to produce mounted CH targets and to deliver at least 250 mounted targets to NRL for the NIKE laser program.

The accomplishment of these subtasks, and the production of the deliverables associated with the subtasks, provided us with many challenges which we have been able to meet successfully. We have delivered more than the required 250 targets for the NIKE laser program and have satisfied all the required demonstrations of fabrication and characterization techniques. One of the specifications on target films is that the surfaces be smooth to better than 10 nanometers. We routinely produce films which have surface smoothness better than 1 nanometer, a quality which has been measured and verified by laboratories outside the program.

In most areas of this task—film production, parts fabrication, characterization, and target assembly—we have satisfied the requirements of the task specification and in some of them we have exceeded those goals by more than an order of magnitude.

1.2.11. SL01 FABRICATION OF FOILS, FILTERS, AND POLYMER

This year we continued the fabrication, characterization and delivery to Sandia National Laboratory (SNL) of foil targets with the production of 321 free-standing gold and aluminum foils. A large box coater was received from KMS and it was brought into operation for the continued production of foil targets in the coming year.

A total of 105 x-ray filters consisting of an aluminum-mylar-aluminum sandwich were fabricated, characterized and delivered to SNL. Part of this task consisted of support to SNL. In response to this requirement, T. Alberts was assigned to the Albuquerque laboratory where he acted as liaison between GA/WJSA and SNL. He fabricated foils and filters with SNL facilities and developed a device for measurement of the attenuation of x-rays through filters.

SNL requested the synthesis and delivery of deuterated parylene. We investigated several routes for the deuteration of parylene and found one that worked but at a rate that was so slow that it was not very practical. However, we did manage to deliver about 25 g of deuterated parylene to SNL.

We also successfully demonstrated the feasibility of fabricating large gold foil spheres by gold coating large polystyrene shells, drilling a small hole in the gold coating and then dissolving out the polystyrene.

1.2.12. UR01 LLE MATERIALS DEVELOPMENT

The task objectives were to synthesize and deliver to UR/LLE 50 g of perdeutero-poly(trimethylsilylstyrene) with specifications for average molecular weight, polydispersity and purity. The task was fulfilled by:

1. Reacting the Grignard reagent of perdeutero-4-chlorostyrene with perdeutero-trimethylsilylchloride to form perdeutero-trimethylsilylstyrene.
2. The silylstyrene monomer produced in step one was subsequently polymerized in three batches.

Two of the resulting three batches of polymer, which met all of the required specifications for purity, average molecular weight and polydispersity, fulfilled the task objectives with delivery of 54.3 g. A third batch (20.6 g), which was also delivered, fell just outside of the molecular weight specification.

1.2.13. CR/LL1 CRYOGENIC LAYER DEVELOPMENT

The goal of the cryogenic layering program is to conceive and demonstrate viable ways to generate uniform cryogenic layers. Selection of the cryogenic fuel layering method is a key decision for the OMEGA Upgrade target insertion system. The choice of layering method affects the capsule type and fabrication method, the scenario for filling and transporting, and whether the fuel layer is formed and characterized inside or outside the target chamber.

For D-T fuel, the method with the greatest experimental database is beta-layering. However, nontritiated fuel, D₂, will most likely be used in a significant fraction of target shots, making the layering choice much more complicated. Several layering methods are being actively pursued at this time: thermal gradient liquid layering at LLNL, plasma discharge at University of Rochester's Laboratory for Laser Energetics (UR/LLE), and joule heating at LLNL. IR heating has been proposed as another method, and initial discussions were underway this past year to do some proof-of-principle experiments. Under 94CR/LL1 we have been looking at some of these alternative methods for producing uniform cryogenic layers. A series of successful clamped shell experiments were conducted where thick liquid layers (~50–80 μm) were levitated. Results of analytical modeling calculations were used to interpret the observed oscillation in the

experiments and the correlation seen between previous liquid layering experiments. The effort for this past year has primarily focused on thermal gradient liquid layering, both experimentally and analytically, with some effort being spent on modeling work in conjunction with the joule heating experiments at LLNL. A next generation series of liquid layering experiments was designed and all of the necessary components have been fabricated.

1.2.14. CR/LA1 BETA LAYERING SUPPORT AT LANL

The beta layering work at LANL during FY94 has concentrated on the development of techniques for improving solid D-T surface layer and image quality as well as improving experimental methodology. Several experiments were performed to attempt smoothing of the D-T solid surface by using an annealing technique in which layer equilibration is performed at temperatures close to the triple point (19.79 K). These experiments have shown that long equilibration times and distortions in the optical path conspire to roughen the solid layer, and prevent a clear view (and consequently an accurate measurement) of the surface of interest. The best solid layer surface finish measured was about 2.0 μm , which compares to a value of 0.32 μm measured on a liquid layer surface, equilibration rates for these experiments were measured to be 11 hours, much longer than previously established rate constants and long enough to allow significant ^3He buildup in the solid layer. The helium accumulation within the solid layer both roughens the surface and distorts the optical path since there is D-T solid on the cell front window. Also, analysis of a sample of our D-T showed it to contain about 4% hydrogen, which could contribute to slowing the equilibration rate and lowering the melting point of the solid layer. This D-T inventory has been replaced with a fresh and uncontaminated supply.

Two new D-T cells have been designed and built in order to provide us with a clearer view of the solid surface and improve equilibration rates to near established values (27 min for fresh D-T). As part of the continuing effort to produce high quality solid surfaces (roughness $\leq 0.5 \mu\text{m rms}$), analytical techniques for improving layer image quality, which translates to improving the analytical accuracy of the surface quality, are also being developed.

1.2.15. CR/LA2 CRYOGENIC LAYERING TECHNOLOGY

This task was centered on two projects. One was the measurement of the flow rate of liquid and gaseous deuterium at low temperatures through fill tubes with micron and sub-micron inside diameters. The other was the development and construction of a 1-dimensional (1-D) spatially resolved Nuclear Magnetic Resonance (NMR) probe for examining fractionation in samples of D-T undergoing beta-layering symmetrization.

An apparatus for measuring flow rates through micro-tubes at low temperatures was designed and the components procured. A CRADA was established with Edward Air Force Base — Phillips Labs for procurement on the micro-tubes. A set of mounted micro-tubes was ordered with an i.d. range of 0.5 μm to 10 μm . When these micro-tubes are received, this experiment will resume.

The 1-D NMR instrument probe suitable for handling tritium was designed and built. It is a doubly tuned (for triton and deuteron frequencies) transmission line probe. Several additions were made to last year's prototype. For handling tritium, primary and secondary containment of the sample are included in the probe itself. To increase spatial resolution, the magnetic field gradient design was changed to a self shielded design. The probe was cooled to low temperatures (as low as 3.8 K). It was tunable to the triton and deuteron frequencies at low temperature. Temperature control was maintained at 20 K with a stability of at least 0.01 K with a liquid helium consumption of 1 liter/hour. Unfortunately, the sample cell developed a leak upon cooling which aborted any attempt to conduct 1-D NMR projection imaging. The sample cell will be repaired in FY95 and the NMR imaging experiment resumed.

1.2.16. CR/UR1 OMEGA-UPGRADE CRYOGENIC TARGET SYSTEM

The upgrade of the OMEGA laser at the UR/LLE to 30 kJ (351 nm) will result in a need for millimeter-size inertial confinement fusion targets filled with D_2 or D-T and maintained at cryogenic temperatures. This mandates a cryogenic target delivery system capable of filling, layering, characterizing and delivering cryogenic targets to the OMEGA Upgrade target chamber. General Atomics has been assigned the task of designing, constructing and testing the cryogenic target delivery system. Figure 1-9 shows the major components of the cryogenic target delivery system.

OMEGA UPGRADE CRYOGENIC TARGET DELIVERY SYSTEM

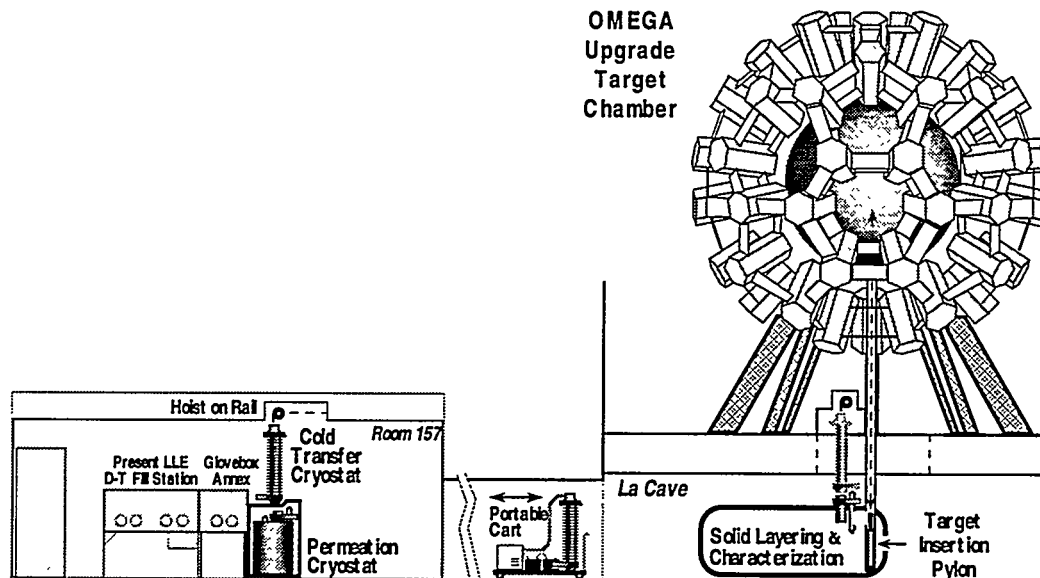


Fig. 1-9. Schematic layout of the target delivery system.

In FY94, we were tasked with engineering activities including the construction of a prototype high pressure fill station capable of filling plastic shells with D_2 gas up to 1,100 atm at room temperature. Major subsystems included a high pressure permeation cryostat, a cryogenic wrench to open the permeation cell and a cold transfer cryostat for cryogenic transport of filled targets. Other activities included conceptual design activities for the target insertion system and providing drawings for modification of the existing UR/LLE D-T fill station.

During FY94 we reassessed the fill station concept, eliminating the need for a separate storage dewar, with improved safety and reduced complexity. We completed designs for the permeation cryostat, the cold transfer cryostat and the cryogenic wrench. Major subsystems ordered included the permeation cell, cryogenic valves, permeation cryostat, and the closed cycle refrigerator for the cold transfer cryostat. All needed components were also ordered. Major construction activity will take place in FY95 when the major subsystems are received from the vendors. Conceptual design of the target insertion system has continued. Two design concepts were developed for target insertion: a cryogenic elevator and a transportable cryostat. Also supplied were modification drawings for the existing UR/LLE D-T fill station.

1.2.17. CR/UR2 CRYOGENIC CHARACTERIZATION AND FOAM DEVELOPMENT

The goal of the cryogenic layering program is to demonstrate viable ways to generate uniform cryogenic layers in a variety of different ICF targets — including glass, plastic and foam. This goal necessarily requires that we must also develop the capability to measure the cryogenic layer uniformity. The efforts taken under direction of task CR/UR2 are summarized below.

Symmetrization of Ice Layers. A program was started this year to investigate the use of an radio frequency (RF) plasma to smooth and symmetrize the ice layer in a cryogenic ICF target. This program was to build on work done by a graduate student at the Institute of Laser Engineering at Osaka University. The student, hired as a postdoctoral fellow at the University of Rochester, designed and built several cavities and demonstrated uniform plasma generation in a room temperature 3 mm o.d. quartz shell filled with H₂. He designed a cryogenic cell, but budgetary pressure at UR/LLE has forced cancellation of this project.

Cryogenic Layer Characterization Development. We have found an optical profiler which is potentially acceptable for the purpose of characterizing the inner surface of a cryogenic target, and have developed a conceptual protocol for using it. In the next year we expect to do analyses to finalize the instrumental configuration and to determine its measurement limitations. Computer programs necessary to analyze the optical profilometry data have also been written.

Foam Shell Development. We developed the capability to make foam shells to the dimensions required by cryogenic targets. Working at and with the staff of LLNL we began with the recipe for TMPT foam with PVP overcoat developed by the Institute of Laser Engineering at Osaka University. After development of a droplet generator, refinements in density matching of phases, and adjustment of initiator activity, we were able to make uniform shells of appropriate dimensions. However, the foam was not sufficiently transparent because of light scattering from the cellular structure. Experiments were then conducted with a resorcinol/formaldehyde (r/f) foam system; it has much smaller cell. In the coming year, experimental programs will explore the feasibility of improvements in the existing foam structure and of making shells using r/f foam.

SECTION 2

COMPOSITE POLYMER CAPSULE DEVELOPMENT AND PRODUCTION

2. COMPOSITE POLYMER CAPSULE DEVELOPMENT AND PRODUCTION

Composite polymer capsule requests sometimes mandated the use of various dopants in the polystyrene mandrel or the ablation layer. These elemental dopants consisted of deuterium (D), chlorine (Cl), titanium (Ti), or silicon (Si) for the mandrel, and germanium (Ge) in the ablation layer. The use of dopants in the mandrel required the custom synthesis of the appropriately doped polystyrene polymer.

2.1. MANDREL FABRICATION

All requested deliveries were fulfilled using low polydispersity polystyrene (plain or doped) mandrels produced by drop-tower. Polystyrene, deuterated polystyrene, chlorinated polystyrene and titanated polystyrene each had different difficulties associated with it as discussed in the following subsections. In addition, bulk, silicon doped, deuterated polystyrene was requested by the University of Rochester.

2.1.1. POLYSTYRENE MANDRELS

While plain polystyrene mandrels pose no particular difficulties, we have continued to make incremental improvements to reduce mandrel-to-mandrel variability. Without these improvements it would have required much more manpower to produce mandrels for the SYMCAP orders (LANL-P07, 8, 9). These three orders required mandrels with tight specifications (445 ± 5 μm o.d. 3 ± 0.3 μm wall) made from both plain polystyrene and a polystyrene/polychlorostyrene blend. A tachometer was installed on the syringe pump which delivers polystyrene solution to the droplet generator. The tachometer reading was correlated with solution flow rate so that the droplet generator can now be set to give the precise amount of polymer required for each mandrel. The problem of polymer solution concentration changing with time due to solvent evaporation was addressed by mixing and storing the solutions in bottles equipped with septa.

2.1.2. DEUTERATED POLYSTYRENE MANDRELS

Deuterated polystyrene was desired for order LANL-PO6. Deuterated styrene (Cambridge Isotope Labs) was polymerized using benzoyl peroxide in benzene, and was precipitated in isopropanol. The initial batch of 33 grams had a $M_w = 138.1$ K, and a polydispersity of 1.79. This material did not make good shells in the drop tower. (A test with narrow polydispersity polystyrene with $M_w = 152$ K also failed to make good shells, showing that M_w near 100 K was necessary.) A second batch (6 g) of deuterated polystyrene was produced, with $M_w = 67$ K and polydispersity of 1.86. The lower molecular weight polymer was used to produce acceptable mandrels for the order. These mandrels are shown in Fig. 2-1.

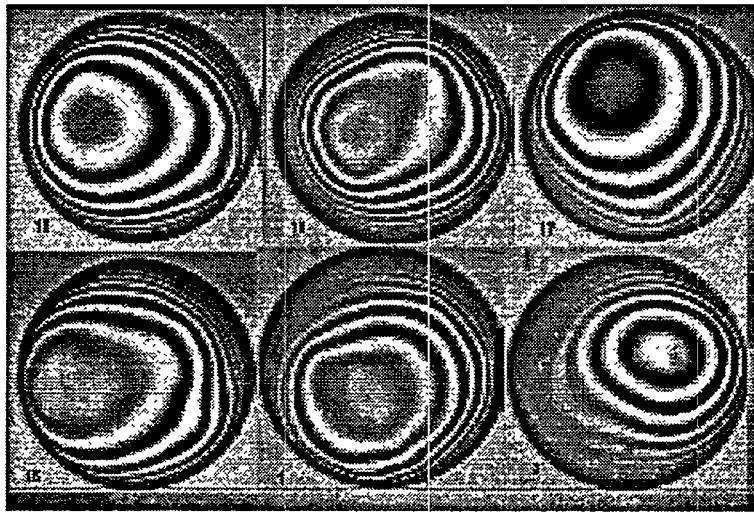


Fig. 2-1. Deuterated polystyrene shells.

2.1.3. CHLORINATED POLYSTYRENE MANDRELS

The polystyrene/polychlorostyrene blend used for chlorine doped mandrels was found by LLNL to produce surfaces with high order modes. Therefore, mandrels were to be made from styrene-chlorostyrene copolymer containing one atom percent chlorine. A custom synthesis of this polymer was ordered from Polysciences, Inc. Although the copolymer met specifications we were unable to produce acceptable mandrels from it. Apparently the copolymer contained a small amount of cross linked material (gel) which led to lumps in the shell walls (Fig. 2-2). We tried to remove the gel by filtration using diatomaceous earth but were unsuccessful in reducing the gel content sufficiently (Fig. 2-3).

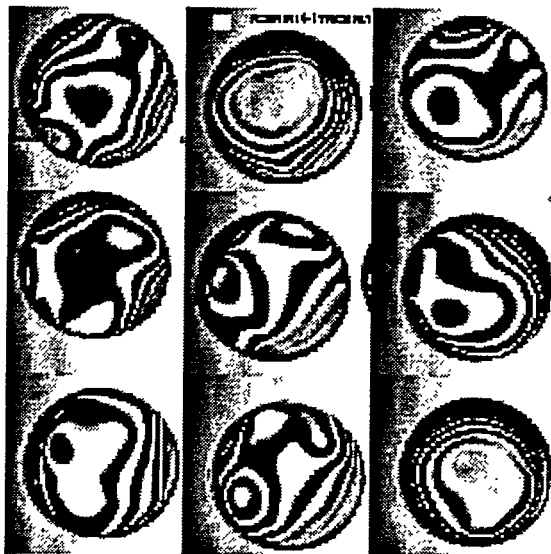


Fig. 2-2. Mandrels from polystyrenes copolymer before gel removal.

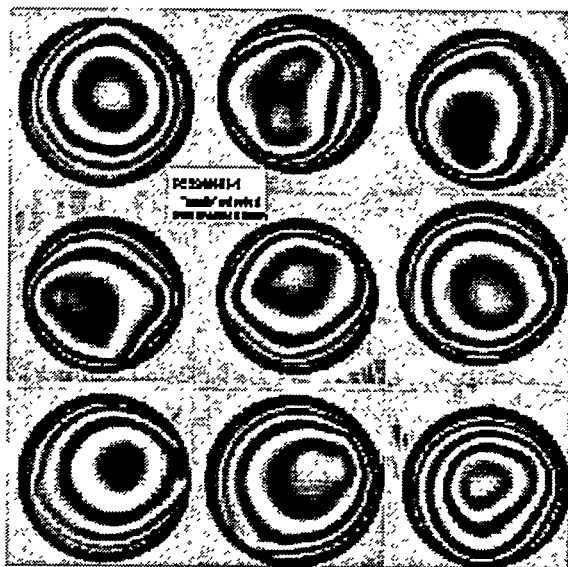


Fig. 2-3. Mandrels from polystyrenes copolymer after gel removal.

GA subsequently produced gel free styrene-chlorostyrene copolymer by addition of 253 cc distilled styrene (Aldrich) and 63 cc distilled 4-chlorostyrene (Aldrich) to 1700 cc of benzene (dried over K), along with 1.32 g of benzoyl peroxide. The solution was heated at 60° C for 24 hours, and the polymer was precipitated in isopropanol. After drying, 54 grams of polymer was obtained, which had a $M_w = 90.9$ K and a polydispersity of 1.73. The polymer was 1.36 ± 0.07 atom percent chlorine, determined by both XRF of mandrels made, as well as combustion analysis. Shells made from the GA copolymer were used to make the delivery (Fig. 2-4).

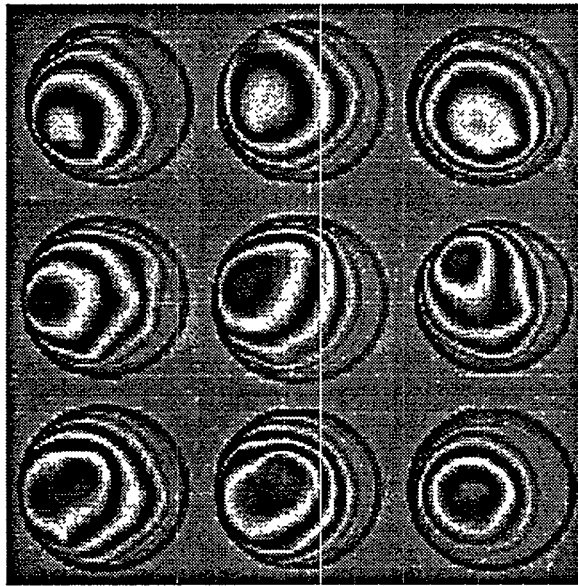


Fig. 2-4. Shells made from GA chlorinated polymer.

2.1.4. TITANATED POLYSTYRENE MANDRELS

A new activity this year was development the capability to produce titanated mandrels doped with titanium (Ti) to 0.06-0.20 atom percent. Two different approaches were used. The first approach was to produce mandrels from chemically bound titanium containing polymers. The second approach was to add nonvolatile soluble Ti compounds to the polystyrene solution usually used for mandrel production.

Titanium-Methacrylate/Styrene Copolymer. Prof. Gary Gray at the University of Alabama-Birmingham (UAB) has been developing a titanium doped copolymer of styrene with methacryloxyethoxy-titanium triisopropoxide. The titanium monomer, manufactured by Gelest, is shown in Fig. 2-5. Examination of the figure shows that the titanium can be lost from the monomer (and likewise the copolymer) through either hydrolysis of the titanium alcoxide linkages or acid cleavage of the ester linkage. Indeed retention of the titanium is a problem with this polymer.

The polymer obtained is typically some 0.80–0.85 mole fraction styrene, and 0.20–0.15 mole fraction methacrylate, based on NMR analysis. The polymer should contain 0.7–0.9 atom percent titanium (% Ti) based on one atom of titanium per methacrylate. Typically a much lower titanium content is observed (0.02-0.12 % Ti). Polymers made at both GA, and at UAB, have been low in titanium. UAB has delivered

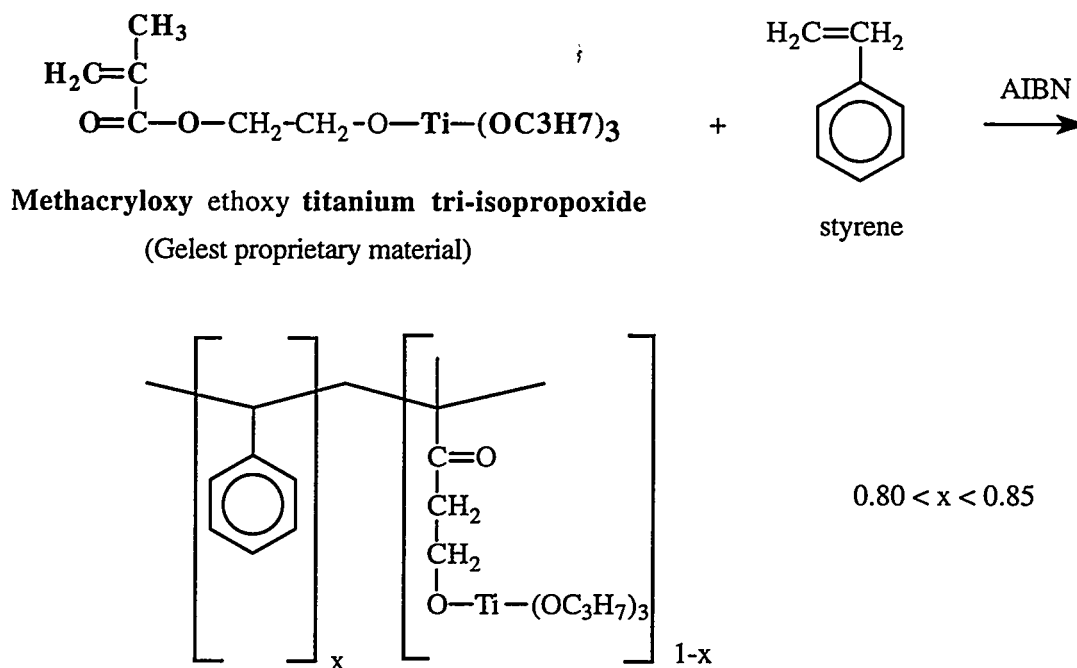


Fig. 2-5. Synthesis of ti-methacrylate/styrene copolymer.

two batches of polymer to GA, which had titanium contents of 0.032 and 0.021 % Ti. Polymer made by GA had titanium contents of 0.039 and 0.034 % Ti. (The desired titanium content is 0.06–0.2 % Ti.) The low titanium content has been the predominant problem with this material, which is discussed below.

At UAB, the polymer is made on a high vacuum glass line, utilizing glass break seals, diffusion pumps, etc. At GA, lacking such elaborate equipment, we have managed to synthesize polymer of the same apparent quality, using a dry box and standard inert atmosphere techniques in standard taper glassware. At GA, the initial work started by handling the monomer in an inert dry box. However, the neat monomer spontaneously solidified within 5 minutes. The monomer is stored in the presence of a polymerization inhibitor, which inhibits only in the presence of oxygen. The absence of oxygen in the dry box leads to the failure of the inhibitor, and the monomer polymerizes. The current practice at GA is to cannulate the monomer into benzene, along with the styrene and AIBN, in glassware full of air. The air is swept out of the system with several freeze-pump-thaw- N_2 backfill cycles, and the polymerization is begun by heating. Both GA and UAB add their solution to dry hexane, to precipitate the polymer over 24 hours. The polymer is redissolved, and reprecipitated, to obtain pure polymer.

The low titanium content has been puzzling to both GA and UAB. There are two obvious sources of the low titanium content. One source could be water present which hydrolyzes the titanium off the methacrylate. GA uses "anhydrous" hexane, while UAB dries their hexane over a molecular sieve. In both cases, the maximum amount of water in the hexane should only remove some 10% of the titanium from the methacrylate (not some 80–90% of the titanium). The second source may be low titanium content in the original monomer. Neither GA nor UAB have regularly checked the neat monomer, which its manufacturer, Gelest, says is pure. (The NMR of one sample showed the absence of the ethoxy group. We suspect they had mislabeled a bottle, as another monomer in their catalog does not have the ethoxy group.) We will be checking each bottle of monomer in the future.

Mandrels were easily produced using both the University of Alabama and GA titanated copolymers. None of the mandrels contained the required minimum 0.06 atom percent titanium.

Titanocene-Styrene Copolymer. Prof. Larry Kool of Boston College has been working with oxygen free titanocene materials, which have thermally stable titanium-carbon bonds. Simple titanium alkyls are not stable at room temperature, which limits the range of titanium species which can be attached to polymers. By attaching a titanocene, specifically a cyclopentadiene-titanium-cycloheptatriene moiety, to styrene, one would have a polymerisable monomer, consisting solely of titanium, carbon and hydrogen. The synthetic route Prof. Kool has taken is shown in Fig. 2-6.

The synthesis of a stable monomer was not successful although several small difficulties were solved along the path. Apparently, the monomer is synthesized, but it tends to polymerize with cross linking (presumably a small amount of divinyl impurity) during isolation which makes the resulting polymer intractable. Prof. Kool has not identified the impurity, nor has he been able to obtain pure monomer or polymer. The material may be intrinsically unstable. The titanium is formally Ti (0), and the Ti is electronically "connected" to the vinyl group. This connection may be leading to the spontaneous polymerization. If a $-\text{CH}_2-$ group were attached between the titanocene and styrene units, the "connection" would be broken, and a more thermally stable monomer may be obtained.

For further information, please contact Dr. B. McQuillan (GA).

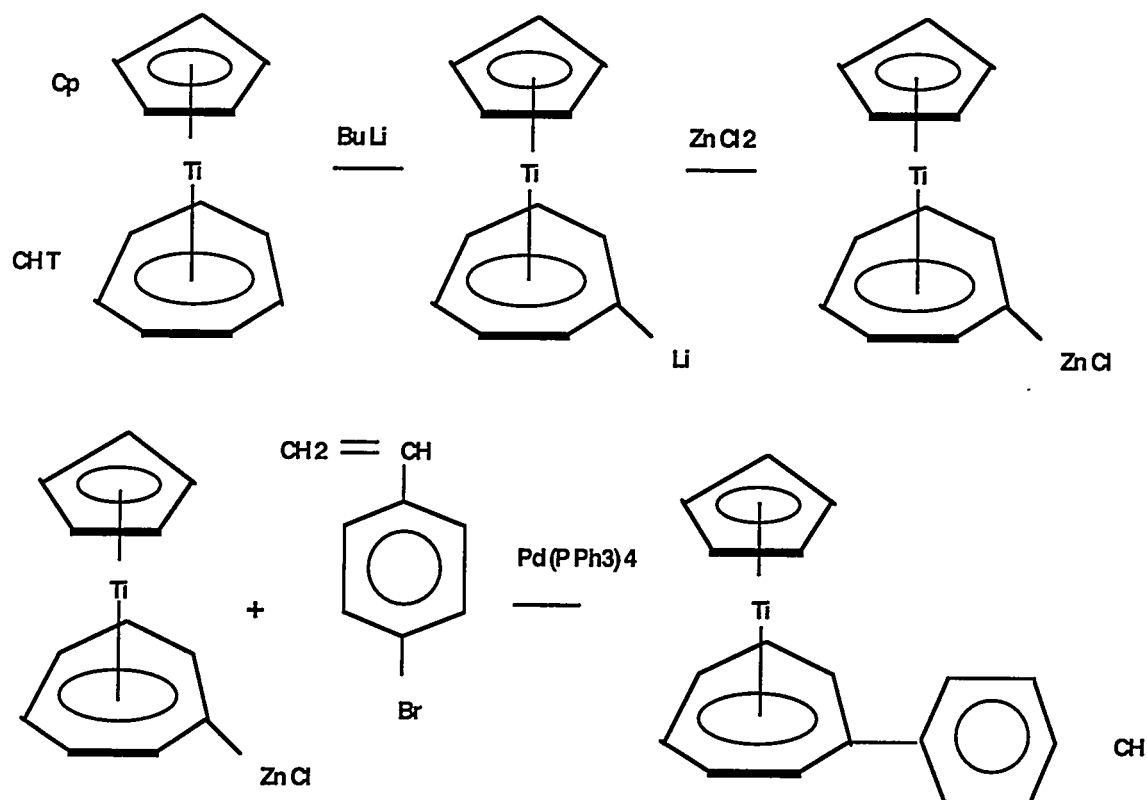


Fig. 2-6. Synthesis of titanocene/styrene copolymer.

Mandrels from Titanium Additives. An alternative method for producing titanium doped mandrels was investigated, the addition of soluble titanium compounds to the polystyrene solution used in the drop tower. The compounds investigated are listed in Table 2-1. Three of the additives produced acceptable mandrels with reasonable yields. The titanium(IV)-bis(ethylacetoacetato diisopropoxide) gave the best results to date and is the prime candidate for use as an alternative source of titanium mandrels. The question of titanium homogeneity in the shell wall is still being investigated. No evidence of inhomogeneity has been observed using EDXS, electron backscatter, x-ray imaging or Auger but the low titanium content in the mandrels limits the sensitivity of these techniques. LLNL is investigating titanium uniformity in these shells using more sensitive techniques.

Table 2-1
Mandrel Production Using Titanium Additives

Compound	Shell Yield	Shell Quality	Titanium Retention
Titanium(IV)-bis(ethylacetoacetato diisopropoxide)	Good	Good	100%
Titanium diisopropoxide bis(2,4-pentanedionate)	Fair	Fair	100%
Titanium (IV) tetrachloride tetrahydrofuran complex	Good	Fair	90%
Titanocene dichloride	Fair	Good	2%
Titanium-2-ethylhexoxide	Fair	Poor ²	NM ³
Titanium n-butoxide	Fair	Poor ²	NM ³
Titanium isobutoxide	Fair	Poor ²	NM ³
Poly(dibutyltitanate)	Zero ¹	NA	NM ³
Cyclopentadienyl cycloheptatrienyl titanium	Poor	Fair	2%
1 Precipitated in bottle before dropping			
2 Mandrels were frosted rather than clear			
3 Not measured but probably 100%			

For further information, please contact Dr. L. Brown (GA).

2.1.5. PERDEUTEROPOLY(TRIMETHYLSILYLSTYRENE) SYNTHESIS

The University of Rochester desired 50 grams of perdeuterated poly(trimethylsilylstyrene) with a polydispersity <2.0 , and a molecular weight $100 \text{ K} < M_w < 250 \text{ K}$. Since the polymer was not commercially available we used the process utilized by Osaka University (Fig. 2-7). All the necessary reagents are commercially available stock items, except for perdeutero-4-chlorostyrene. Perdeutero-4-chlorostyrene (+98%) was obtained from Cambridge Isotope Lab as a custom synthesis. Products were characterized by gas chromatography – mass spectroscopy (GC-MS).

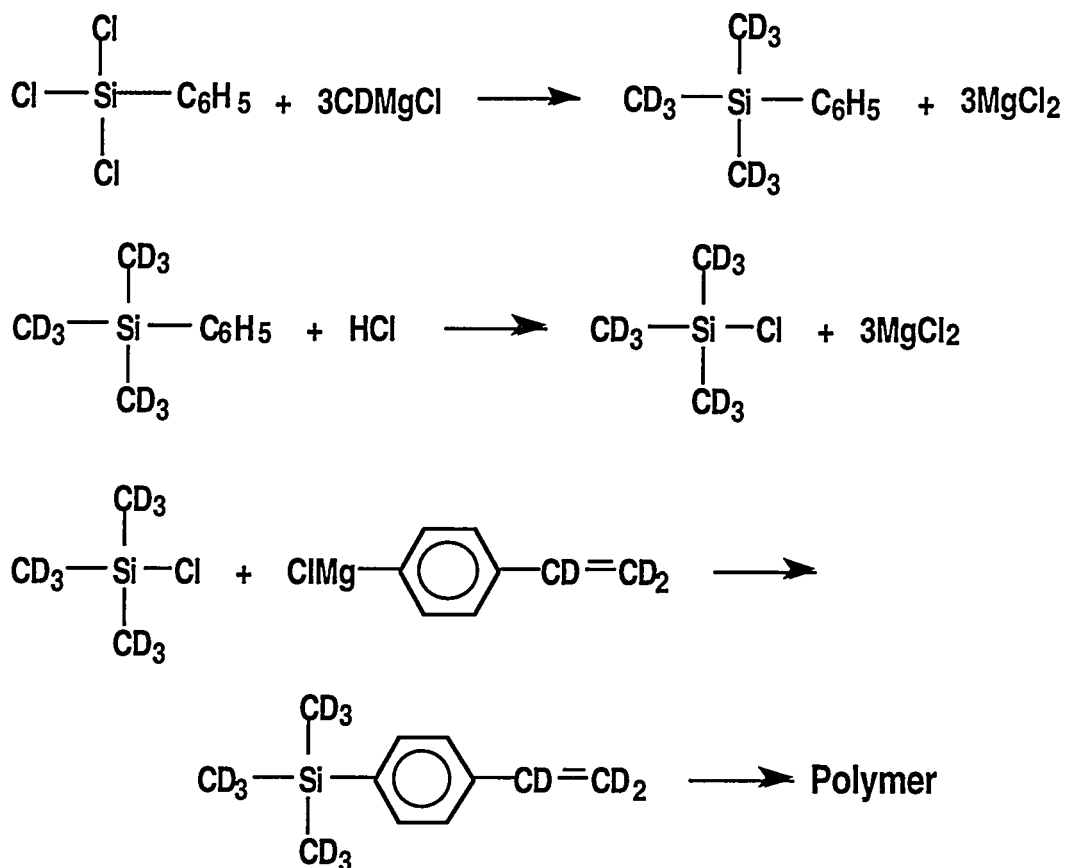


Fig. 2-7. Steps in perdeuterated poly(trimethylsilylstyrene) synthesis.

The reaction steps in Fig. 2-7 were performed under nitrogen. Evolved heat was removed by ice baths. Intermediate, nonhalogenated materials were washed with excess water to remove salts, and dried over calcium chloride. The trimethylsilyl chloride product was saturated with hydrogen chloride. The presence of any hydrogen chloride in

the material would be detrimental to the next reaction as it would react with the Grignard of 4-chlorostyrene, placing a hydrogen atom in the 4-position, thus decreasing the deuterium and silicon content. The vast majority of the hydrogen chloride in the trimethylsilyl was removed by slow gas evolution through a calcium chloride drying tube attached to the flask in a fume hood over several days. The products benzene and trimethylsilyl chloride were not separated, but were used together in the final Grignard reaction. Ten ml of D₂O was used to kill any excess Grignard in the final step, forming perdeuterostyrene (C₈D₈), so as to not introduce any hydrogen to form C₈D₇H. The final trimethylsilylstyrene was distilled under vacuum to remove it from impurity divinyl materials which would cross link the polymer. Ultimately 200 grams of +99% pure trimethylsilylstyrene were produced with 75.7% yield.

Polymerization was performed using benzoyl peroxide in benzene, at about 50°C, aiming for about 30%–40% yield. The polymer solution was filtered using diatomaceous earth to remove gel, then precipitated in isopropanol. Remaining monomer was recovered from the isopropanol and reused to make another batch. Ultimately, three batches of polymer were made, and their molecular weights were determined by GPC. The bulk density of this deuterated polymer was determined by gas pycnometry. The level of deuteration is estimated to be $99 \pm 1\%$, based on GC-MS and the purity of starting reagents. Elemental analysis was performed by Galbraith for chlorine impurity as well as carbon, hydrogen and silicon. The residual chlorine in the polymer, from 4-chlorostyrene, amounted to 1 chlorine atom for every 62000 silicon atoms. A comparison of the specifications with the results of the three batches delivered are given in Table 2-2.

Table 2-2
Deuterated Poly(trimethylsilylstyrene) Delivered to LLE

Specification	Batch 1	Batch 2	Batch 3
50 grams	44.3	9.9	20.6
$100 \text{ K} < M_w < 250 \text{ K}$	165.9 K	154 K	74.4 K
Polydispersity < 2	1.79	1.92	1.72
Density of polymer 1.0706 ± 0.0003			
Chloride content 248 ppm by weight			

For additional information, please contact Dr. B. McQuillan (GA).

2.2. COATINGS FABRICATION

There generally are two layers of coatings applied to the bare polystyrene mandrels. The first is a gas barrier layer which consists of polyvinyl alcohol and the second is an ablation layer which consists of glow discharge polymer.

2.2.1. POLYVINYL ALCOHOL COATINGS

An objective in the first quarter of FY94 was to increase the reliability of our PVA coating process. We wrote an operating procedure to document our standard coating operation. After internal review and approvals, the document was released in January [Ref. 2-1]. Operation using this procedure was the baseline used to evaluate further improvements to the process.

As the year progressed we made numerous improvements to the PVA coating process. Some of these are listed in Table 2-3. One equipment improvement, the new PVA fill/injection device, is shown in Fig. 2-8. This new device both reduces debris and minimizes the forces on the mandrels during PVA fill and injection. Using this device we no longer lose complete runs due to accidental crushing of the mandrels in the capillaries.

Table 2-3
PVA Process Improvements

Procedure Modification	Effect	Result
Mass flow control on stripping gas	Reproducible stripping	Improved yield
Humidity control on stripping gas	Reproducible stripping	Improved yield
Elimination of evacuation/Ar fill	Minimize shell sticking	Reduced strings
Dry sieving	Cleaner mandrels	Reduced debris
New PVA fill/injection device	Minimize PVA flakes Minimize forces	Reduced debris Reduced breakage
Plug capillaries while grinding tips	Cleaner capillaries	Reduced debris
Ultrasonic cleaning of capillaries	Cleaner capillaries	Reduced debris
Ultrasonic cleaning of stop wires	Cleaner capillaries	Reduced debris

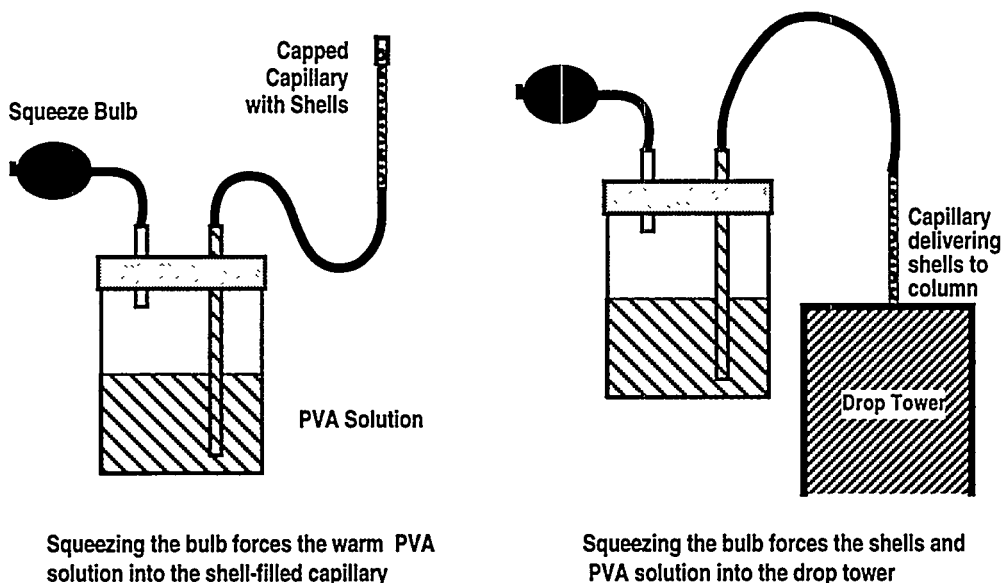


Fig. 2-8. New PVA fill/injection device decreases debris.

As we have introduced these improvements our yields have increased. Figure 2-9 compares the yields for FY93, October through March 1994 and July through September 1994.

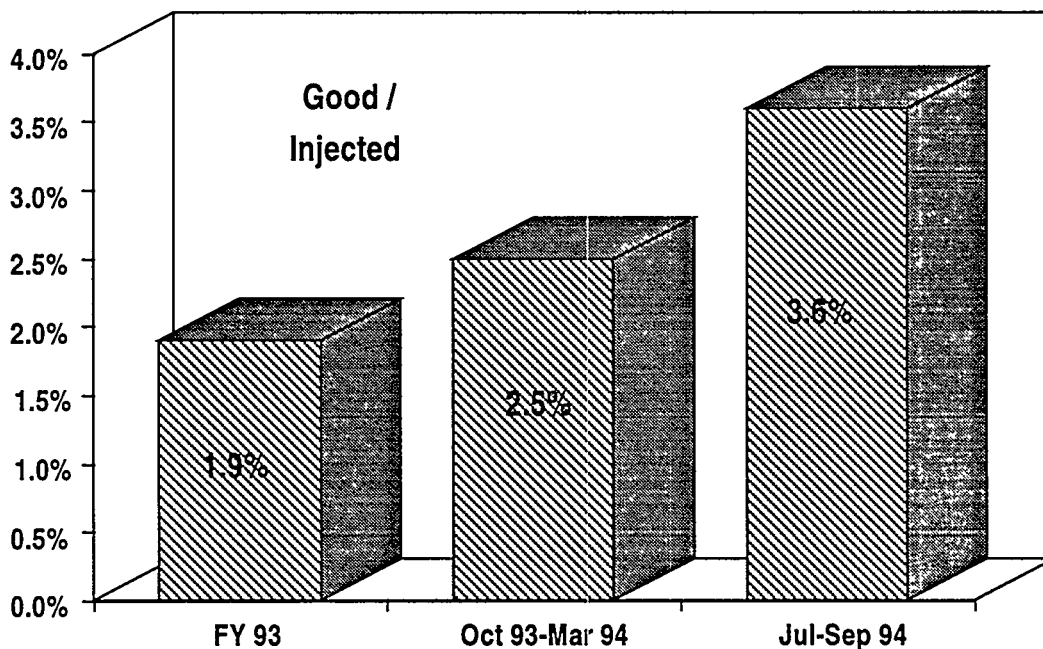


Fig. 2-9. Process yields and quality of PVA coatings have increased.

For further information, please contact J. Gibson or D. Husband (GA).

2.2.2. GDP COATINGS

The GDP Coatings Group coated capsule batches for twelve orders this year. The yield of the GDP process was such that we never had to repeat a coating run to fill an order, although we sometimes split a batch of shells between two coaters and coated them simultaneously. The number of capsules delivered met or often exceeded the number requested, falling short in only one instance, in which case the shortfall was not due to a coating problem. All deliveries were within the specified tolerances for wall thickness and surface finish. Furthermore, our ability to control the coating rate has allowed the wall thickness tolerance to be tightened from ± 5 to ± 3 or even ± 2 μm .

In-situ Thickness Measurement Diagnostic. We continued development of the technique for measuring coating thickness without removing capsules from the coater during the first half of this year. Although this method is within 10% of the measurements as made in the characterization lab, this technique has not been adopted for use in deliverable runs. The vacuum chamber is opened only once for measurements done in the characterization lab which are more accurate. The effort required for both the in-situ and characterization lab measurements is about the same. Furthermore, once thickness measurements have been made, shells can be replaced in the coater for additional coating without adversely affecting the surface finish of the batch. As a result of the above observations, we continue to use the characterization lab measurements for deliverables.

The in-situ video diagnostic can be used for development work and to monitor the coating process. For example, in-situ measurements show how the coating thickness changes during a run. At the start of a run, the shells tend to reside high in the coating pan, where the coating rate is lower. As they become heavier they move down into the center of the pan where the coating rate is highest. We wanted to know the effect this had on coating rate. Video measurements revealed that the coating rate is slightly lower at the beginning of a run, and that the coating rate is fairly constant after about 100 hours of coating, as can be seen from Fig. 2-10.

The measurements for Fig. 2-10 were made using witness shells to avoid the inaccuracies involved in using batch averages. This brings up a question as to whether a witness shell, which has a larger or smaller diameter than the batch, coats at the same rate as the shells in the batch, since earlier studies with stalk-mounted shells had shown that

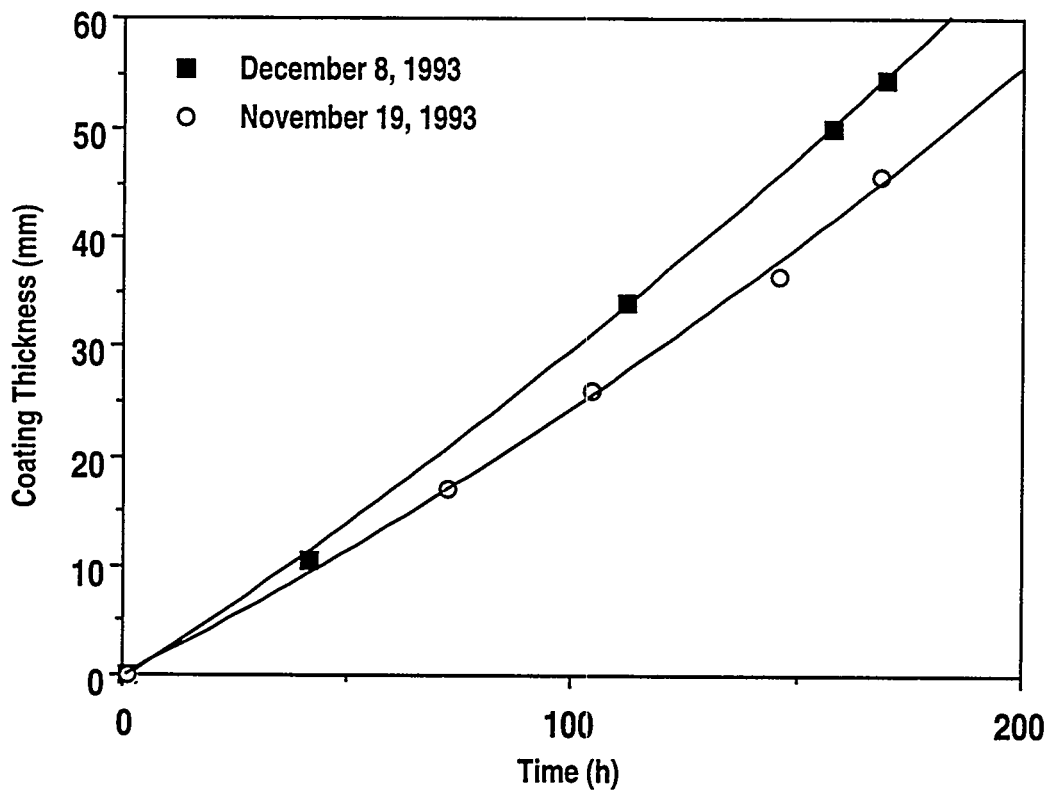


Fig. 2-10. Coating thickness as a function of time elapsed for two coating runs.

coating rate decreases with increasing substrate diameter [Ref. 2-2]. We therefore did a run with two witness shells, one larger and one smaller than the batch. The result was that both witness shells coated faster than the batch, which was surprising, at first.

To obtain the different diameters, the witness shells were obtained from a different polystyrene tower run than the other shells in the batch. The witness shells had thicker walls and were therefore heavier than the other shells in the coating pan. We found that, for shells in the diameter range that we usually coat, substrate mass affects the coating rate substantially more than diameter. Figure 2-11 shows graphs of coating rate vs. diameter and coating rate vs. mass. The data points on the top graph show little relationship between coating rate and diameter, while the lower graph, for the same shells, shows coating rate increasing with increasing substrate mass.

Germanium-Doped Coatings. This year we began to produce capsules with germanium (Ge) -doped coatings. We do this by adding two to four hundredths of an sccm of tetramethylgermanium to the usual CH coating mixture of 0.135 sccm of

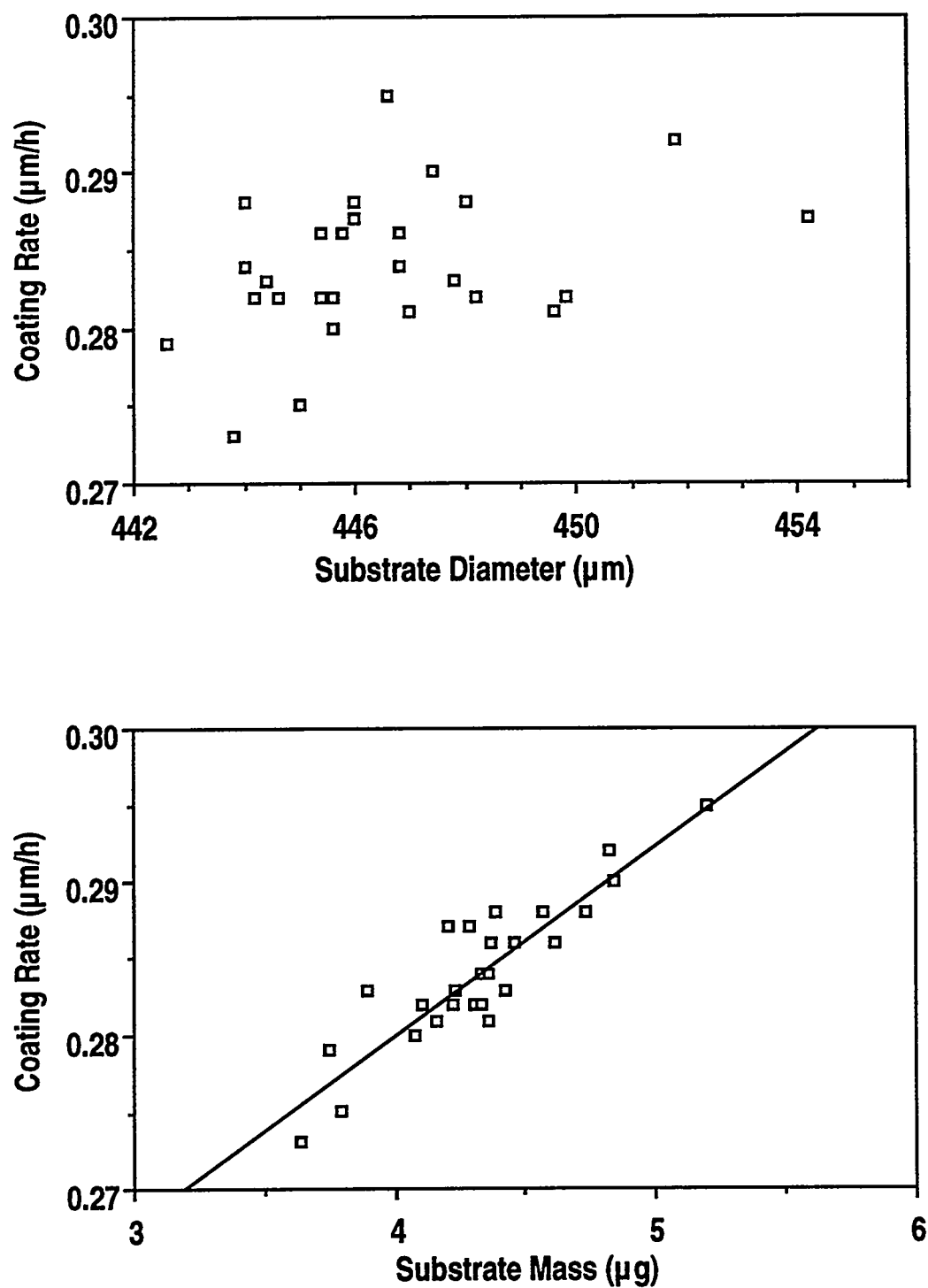


Fig. 2-11. Graphs of coating rate vs. substrate diameter (top) and vs. substrate mass (bottom) for the same set of shells.

trans-2-butene and 10 sccm hydrogen. We have completed four shakedown runs and have filled one order calling for Ge doped shells. Data from some of these coating runs show that increasing the germanium concentration increases the coating rate, while the surface finish remains well within specifications. As you can see from the graph (Fig. 2-12), the surface finish becomes somewhat rougher as the Ge concentration increases, but remains well within the 100 nm RMS tolerance. The capsule coating rate increases from about 0.3 $\mu\text{m/h}$ to about 0.5 $\mu\text{m/h}$ when the Ge concentration in the coating is increased from 0 to almost 2 At%.

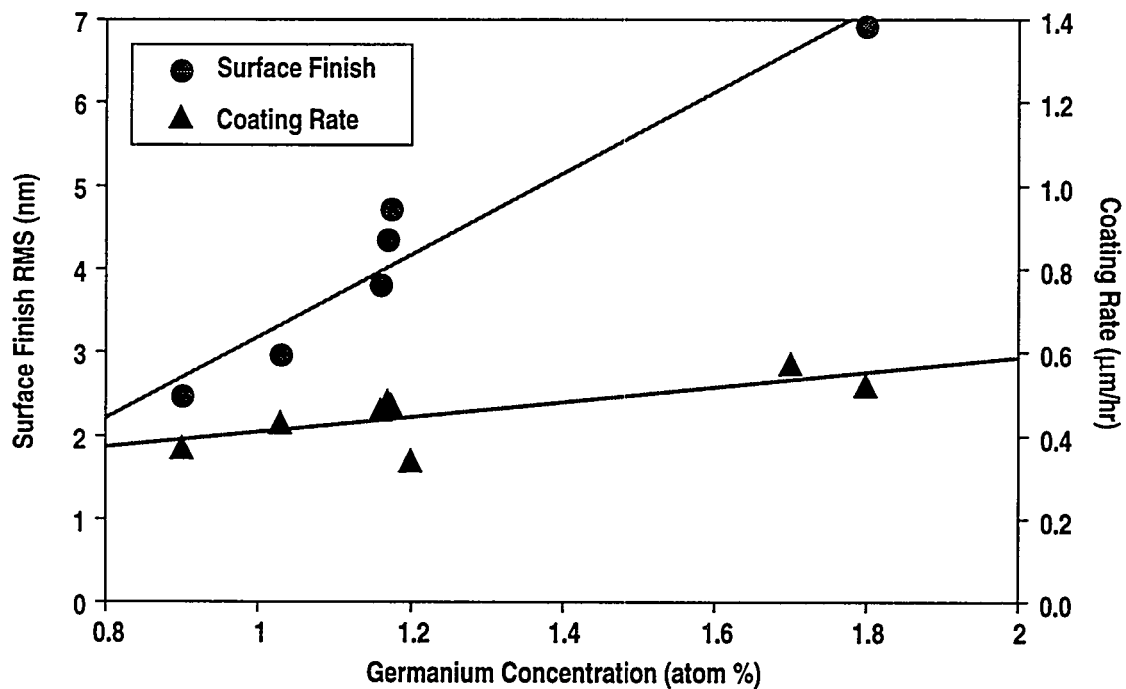


Fig. 2-12. The effect of Ge concentration on capsule surface finish and coating rate for coatings 40–60 mm thick.

For further information, please contact J. Ankney (GA).

2.3. CHARACTERIZATION

Two additional characterization techniques were added this year which have significantly added to our characterization capabilities.

2.3.1. X-RADIOGRAPHY FOR PRECISE SHELL MEASUREMENTS

We have made substantial improvements in the understanding and use of this technique since first describing it in the 1993 Annual Report. Our system consists of a version of *NIH Image* which has an added radial averaging module. This module creates data files used by *Excel* spreadsheets. Both the module and the spreadsheets have been substantially improved in the last year. Used together, they can detect interfaces between dissimilar plastic layers and then locate those interfaces with an uncertainty of $\approx 0.2\mu\text{m}$.

The improved capabilities arise from modifications in five areas: 1) modification of the x-ray spectrum to optimize image contrast, 2) a more sophisticated interpolation scheme for calculating radial data, 3) 2nd derivatives to find interface locations, 4) separation of the radius and layer thickness data, and 5) an *Excel* macro which automates peak finding.

The process begins with an x-radiograph recorded on high resolution x-ray film. The film graininess and penumbral blurring are each $<0.5\mu\text{m}$. The image is digitized using a computer to average 16 8-bit images from a microscope CCD camera [Fig. 2-13(a)]. The film is coincident with the focal plane of the camera to within $\approx 5\mu\text{m}$. The shell image is typically some 400 pixels across. For our standard 1/2 mm diameter shells, that means $\approx 1.2\mu\text{m}$ pixels.

The surfaces and interfaces are identified as cusps in the radial intensity function. Any single line-out from this picture would locate such cusps to within 1 pixel, if the noise were not too great [Fig. 2-13(b)]. The circumference of the image contains ≈ 1200 pixels, which correspond to 1200 independent measurements of each edge.

Radial averaging puts these lineouts together so that the information about the edge adds coherently, and the noise adds incoherently. That average reduces the noise by about $35\times (\sqrt{1200})$ and produces a very smooth radial intensity function [Fig. 2-14(a)]. The 2nd radial derivative of this function is more useful in locating interfaces [Fig. 2-14(b)]; in that case, peaks define the location of interfaces and surfaces.

X-ray Spectrum Modification. The differing x-ray absorption of shell layers imposes a sharp break in slope on the smoothly varying radial intensity (this cusp appears as a peak in second radial derivative). The visibility of this cusp is maximized for an x-ray absorption length equal to the material path length. Unfortunately, the x-ray spectrum

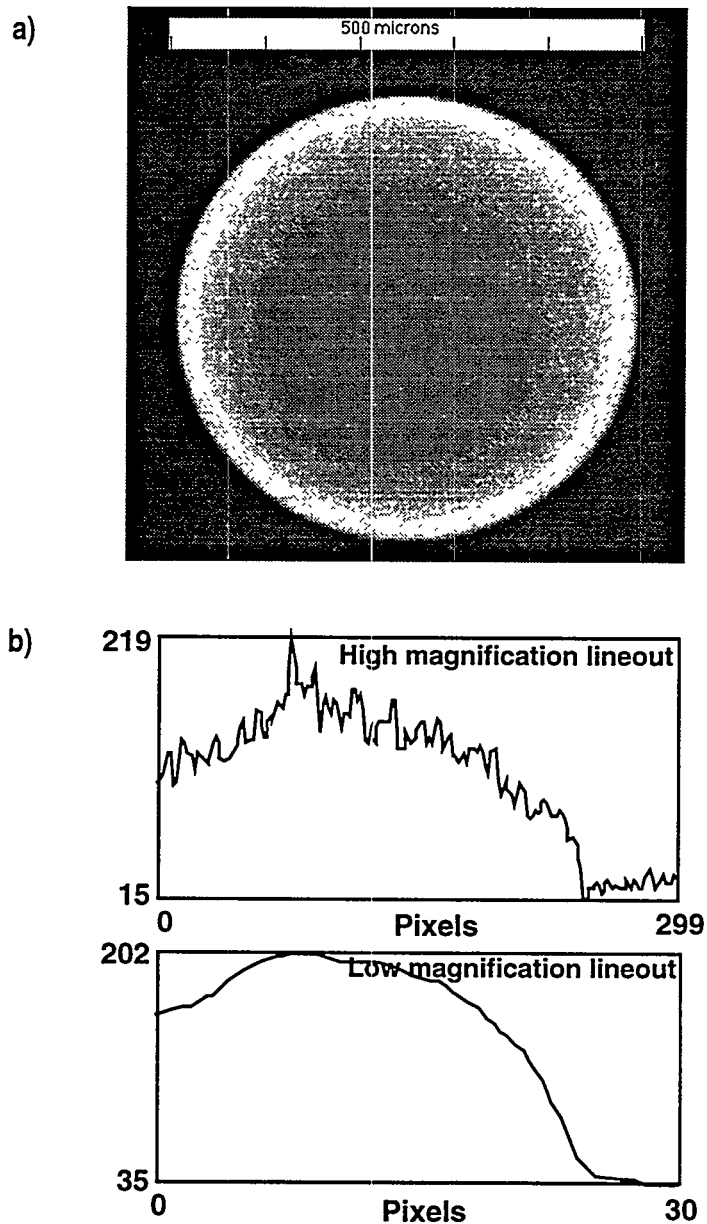


Fig. 2-13. (a) X-radiograph of typical ICF shell. (b) Radial intensity from a single line-out from a at normal magnification and at 10X magnification (the latter resolves individual film grains). (c) 360° averaged radial intensity and 2nd radial derivative for the image in (a)

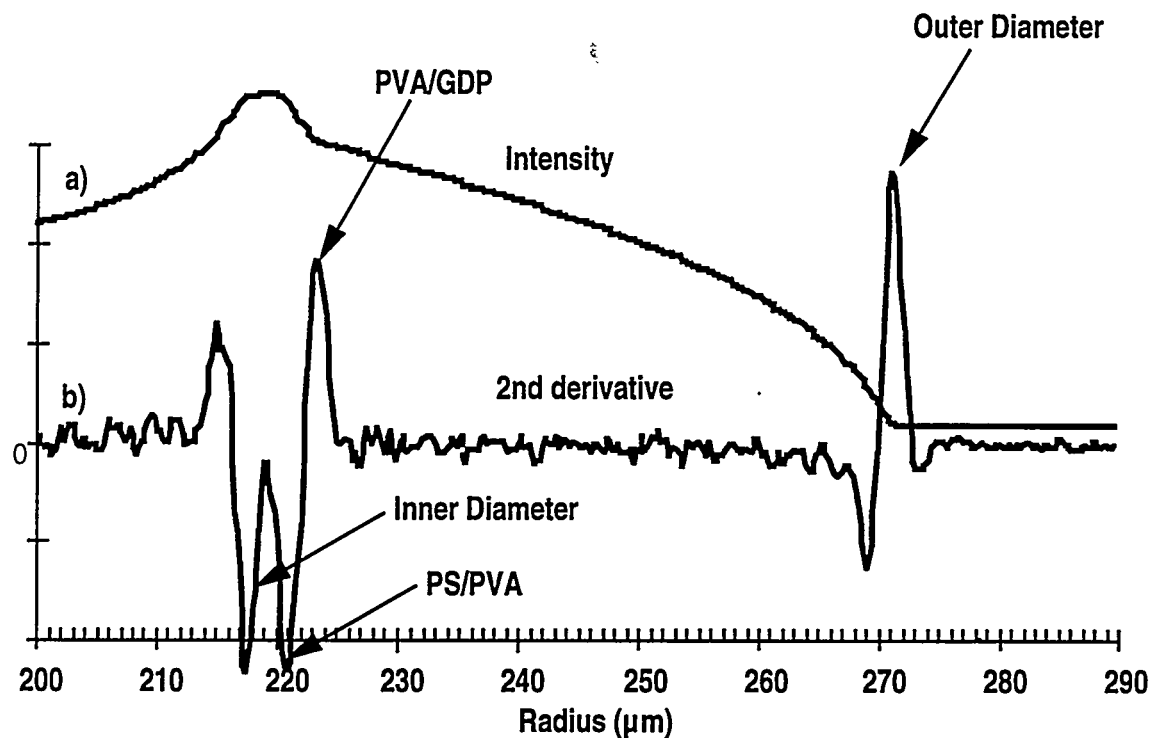


Fig. 2-14. 360° averaged (a) radial intensity and (b) 2nd radial derivative for the image in Fig. 2-13(a).

from our tungsten anode tube is better suited to study of the bare mandrels than the finished target (Fig. 2-15). We use a plastic filter to modify the output spectrum. That improves the contrast by $\approx 4\times$ but results in a four hour exposure time.

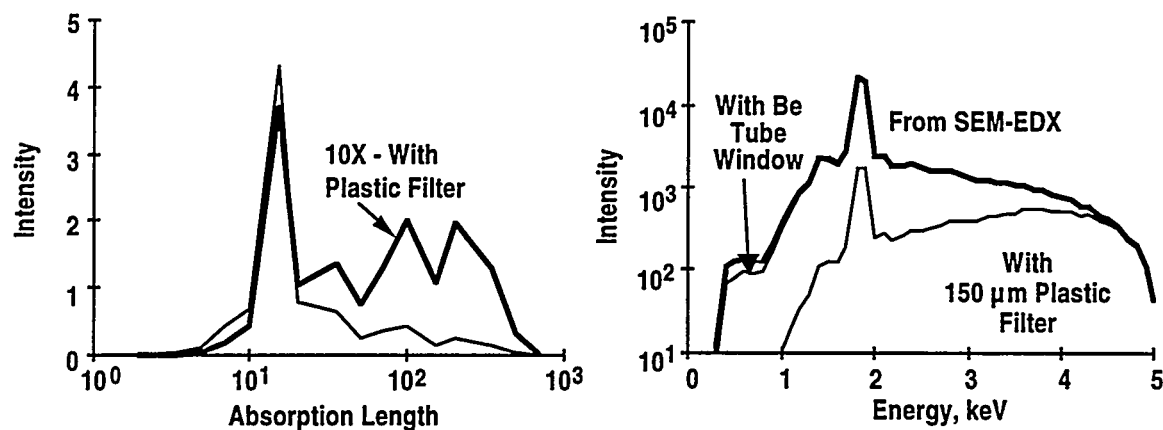


Fig. 2-15. X-ray spectrum from W-anode x-ray tube operated at 5 keV graphed (a) vs. x-ray energy and (b) vs. CH absorption length. Unmodified $\approx 80\%$ of the x-rays have a 1020 μm absorption length.

Data Interpolation. Radial intensity information is generated by interpolating between the rectangular lattice of data points which make up the digitized shell image. We use a two-dimensional cubic interpolation (spline fit) using 12 data points surrounding the point in question. We use these spline fits to calculate the intensity value at a set of points along radii, and then calculate a second derivative by taking differences of the values at those points.

Second Derivative Peak Location. A shell interface (or surface) shows up in an X-radiograph image as a discontinuity in the slope of the image brightness vs radial distance. Assuming a linear response (x-ray absorption and film density proportional to absorber path length) the film density, $D(r)$, should vary near a layer interface like

$$D(r) = A \left(1 + \Delta B R \sqrt{1 - r^2/R^2} \right) \quad r \leq R \quad (1)$$

where R = interface radius,

$A \propto$ x-ray exposure,

$$\Delta B = B_{r < R} - B_{r > R}$$

$B \propto$ plastic opacity on the indicated side of the interface,

and $D(r) \propto$ the log of the brightness of the digitized image,

The edges are easier to locate if we take the second derivative of Eq. (1):

$$\frac{d^2 D(r)}{dr^2} = -\frac{\Delta B}{R} \left(1 - r^2/R^2 \right)^{-3/2} \quad r < R \quad (2a)$$

$$= \Delta B R \delta(r - R) \quad r = R \quad (2b)$$

which converts the slope discontinuities into an asymmetric pair of opposed peaks. The edge is centered under the major peak. The sign of the peak depends on which side of the interface contains the stronger absorber.

Separation of Shell Radius and Layer Thickness. The width of the peaks resulting from the 360° radial average is determined by the variation in shell radius as a function of angle. Large variations spread out the peaks and reduce their height. For many shells, the layer thicknesses are much more uniform than the radius, so the peak intensity can be substantially enhanced if the radial variation is removed from the data [Fig. 2-16(b),(c)].

That is possible because the peak corresponding to the outside edge is very large, and can be reliably detected without angular averaging. In order to separate shell radius and layer thickness, the National Institute of Health (*NIH*) *Image* analysis module makes two passes through the image data. In the first, it locates and records the outer edge of the image for every angle. In the second, it offsets every radius such that the outer edges exactly line up. Then the rectangular plot on axes of radial distance and angle more clearly shows variation in thickness of the layers [Fig.16(c)]. The 360° average of that data exhibits increased signal to noise because the radial variations have been removed. The angular dependence in the radius is included separately in the exported data file.

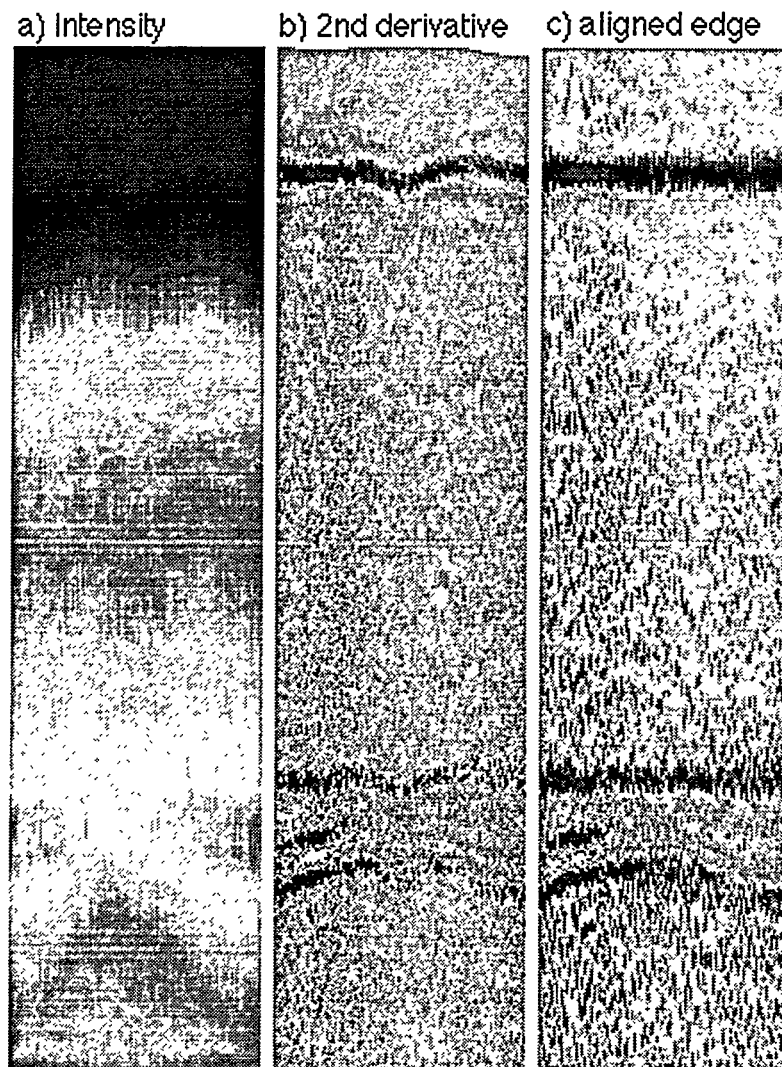


Fig. 2-16. Rectangular plots of the 2nd radial derivative of intensity on axes of radial distance (200–280 μm — vertical) and angle (0–360° — horizontal). They show 100 radial lines, each the average of 30 radii.

Automatic Peak Finding. For analysis, the data file is pasted into an *Excel* spreadsheet and associated macro. The shell radius vs angle is plotted on one graph, and the 2nd radial derivative of intensity vs radius is plotted on the other. A button on the chart activates a peak finding routine which finds the peaks, locates their center by fitting parabolas to them, and calculates the average radius and layer thicknesses from their location. This spreadsheet has switches which allow the user to over-ride the peak finding routine or to give hints to it if the data is poor.

Finally, the macro accumulates the radius and layer thickness data for a series of shells into a single summary spreadsheet.

Procedure Acceptance. The modified *NIH Image*, and the *Excel* spreadsheet have been released for use. Copies have been requested by LANL and LLNL.

For further information contact Dr. R.B. Stephens (GA).

2.3.2. X-RAY FLUORESCENCE

Our x-ray fluorescence instrument (XRF) is currently calibrated for quantitative measurement of all current dopants in ICF capsules. These include: chlorine and titanium in polystyrene mandrels; argon in polymer and glass shells; and germanium in glow discharge polymer coated shells.

All of the calibrations have been achieved using GA manufactured standards. Final calibration values have been approved and accepted by LLNL. XRF calibration for Ar was achieved initially by using a fill-tube mounted shell provided by LLNL. Refinements in the Ar calibration were achieved by using permeation filled shells. The subsequent use of permeation filled shells allowed us to readily determine and correct for the effects of various total wall thicknesses on the Ar pressure by allowing us to use capsules with a wide variety of wall thicknesses. Calibration of the XRF for Cl in polystyrene mandrels was achieved by using shells manufactured from chlorinated polystyrene which had been analyzed in duplicate by combustion analysis at Galbraith Labs. Calibration of the XRF for the remaining dopants was achieved by correlating the analyte signal of interest (Ge or Ti) to the Cl count ratio obtained in a thin film of material which has a precisely known A/Cl elemental ratio (A = analyte). A neat diphenylgermanium dichloride film (~1 μm thick) was used for the Ge calibration. A thin Ti-doped polystyrene film cast from

a dilute bis(cyclopentadienyl)titanium dichloride/polystyrene solution in toluene was utilized for the Ti calibration.

Calibration of the XRF for Ge, detailed in [Ref. 2-3] and summarized here, was the most difficult of the current elements. The following effects were responsible for this attribute: the rather large variation in the desired ablator wall thicknesses; the broad range in possible Ge dopant levels (~0.5-3 At%); the substantial x-ray absorbing power of Ge; and the volumetric expansion of the Ge doped ablator layer as a function of Ge concentration. Calibration of the XRF for Ge was accomplished by starting with a group of fully characterized plain polystyrene shells which were matched for wall thickness,. The shells were put into the Ge/GDP coater and coated with ~5 μm of Ge-doped GDP. Two of these shells were removed and analyzed for Ge. The remaining shells were coated with an additional ~8 μm of Ge/GDP. Two more shells were removed and analyzed. In addition, two new polystyrene shells were added to the shells still remaining in the coating pan to serve as controls to confirm that the concentration of Ge in the GDP was not changing as a function of time. Coating of the shells in the pan was resumed. After each subsequent addition of about 7 or 8 μm 's of Ge/GDP, four shells would be removed from the pan for analysis and two plain polystyrene shells would be added to the pan. Of the four shells removed, two had the full allotment of Ge/GDP coating; and the other two were the control shells which had been added at the previous breakpoint.

The XRF data obtained from the Ge calibration experiment, after converting to a non-absorbance corrected, non-expansion corrected At% Ge in the GDP coating, was fit to the general equation for x-ray absorption:

$$I = I_0 e^{-kt}$$

where I is the observed concentration; I_0 is the actual concentration; k includes terms for mass absorption coefficients and elemental density; and t is the GDP thickness. I_0 and k were determined from a plot of $\ln(I)$ vs t . For a GDP coating doped to about 1.7 At% Ge it was estimated that no more than 10% of the x-ray attenuation was due to the carbon, the remainder was due to the Ge. Thus the k term was split into two parts, 90% k_1 (Ge) and 10% k_2 (C). It is assumed for Ge concentrations of current interest that k_2 is a constant.

After correcting the observed At% Ge for attenuation effects, the At% Ge was then corrected for volumetric expansion of the GDP, caused by increased Ge doping levels,

according to the method provided by LLNL and condensed to the equation $At\%_{actual} = At\%_{abcor} / (1 - 0.0371 * At\%_{abcor})$. Figure. 2-17 shows the plot of the uncorrected and corresponding corrected At% Ge data for the shells used in this calibration experiment. Table 2-4 shows the agreement in results obtained on a series of Ge doped shells which have been analyzed by both GA and by LLNL. This empirically derived formula for determining the actual At% Ge has subsequently been entered into an Excel spreadsheet in order to automate future At% Ge calculations.

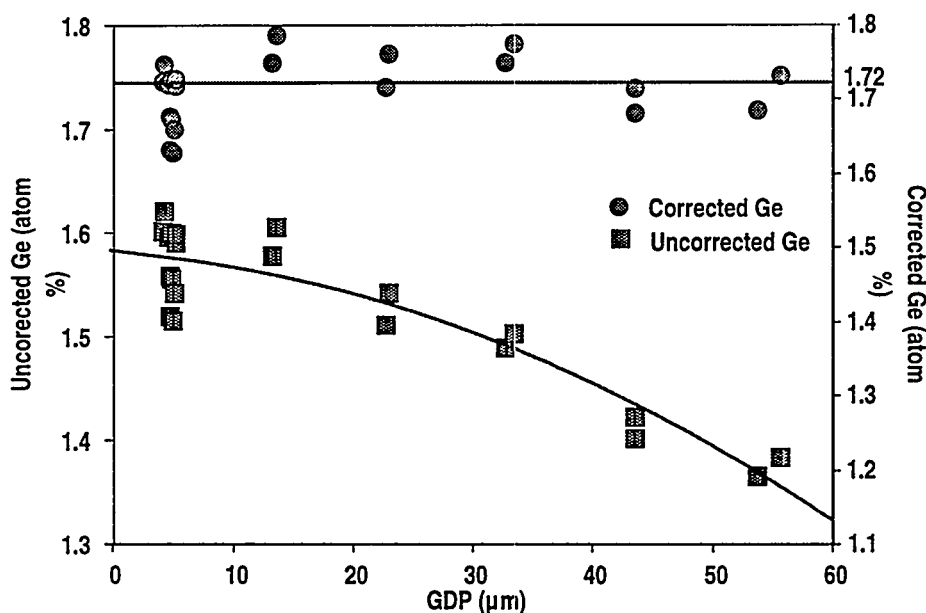


Fig. 2-17. The uncorrected and corresponding corrected AT% Ge as a function of GDP wall thickness.

Table 2-4
Agreement Between GA and LLNL for At% Ge in Composite Capsules is Good

Sample	o.d. (μm)	GDP (μm)	AT% Ge (at GA)	(at LLNL)	% diff
HRA354-3	529	38.3	2.47	2.62	-6.0
HRA342-4	534	33.1	1.35	1.33	1.8
HRA342-3	532	34.0	1.29	1.25	2.8
HRA353-4	473	10.3	2.60	2.44	6.6
HRA353-3	468	10.3	2.46	2.39	3.1
HRA353-2	473	10.3	2.53	2.41	5.1
HRA353-1	463	10.3	2.66	2.56	4.1

Although the Ge calibration agreement between GA and LLNL is fairly good, there remains a small discrepancy. LLNL scientists are satisfied with this level of agreement but we hope to improve our calibration agreement by performing a short series of carefully planned experiments in the coming year.

For further information, please contact Dr. M. Hoppe (GA).

2.4. DIAGNOSTIC GAS PERMEATION

We have developed a procedure for permeating diagnostic gases into GDP-coated composite polymer capsules.

2.4.1. BACKGROUND

In the 1980's, LLNL scientists successfully developed a technique for filling composite polymer capsules with argon early in the capsule fabrication process [Ref. 2-4]. This technique consisted of loading bare polystyrene shells into a capillary tube, evacuating air from the tube, backfilling the shells with the desired amount of diagnostic gas (often mixed with D₂ to maintain a specific partial pressure), covering the shells with an aqueous solution of polyvinyl alcohol, then injecting the shells into a heated tower where the PVA would solidify and entrap the argon in the shells. The PVA-coated shells would then receive a thick (typically several tens of microns) CH-coating in a glow discharge polymerization coater.

This previous diagnostic gas filling technique had several shortcomings. Among them was the difficulty in stockpiling PVA-coated shells having the assortment of argon fill pressures requested by experimentalists. In addition, the maximum argon fill pressure achievable with the previous technique was about 0.8 atmospheres. A partial vacuum had to be maintained in order to allow the aqueous PVA solution to fill the shell-loaded capillary. Another shortcoming of the original technique was that the argon pressure from shell to shell varied by up to 20% due to argon out-gassing from the shells before the PVA had solidified (i.e., in the capillary and/or tower). Finally, the possibility existed that argon could permeate from the shell during the GDP-coating process if deposition conditions caused the shells to become overly hot.

2.4.2. PERMEATION FILL EXPERIMENTS

We built a simple apparatus to permeate-fill composite polymer capsules (Fig. 2-18). This apparatus used off-the-shelf fittings that could withstand temperatures approaching 200°C while pressurized to 60 atmospheres. We used both PVA- and GDP/PVA-coated polystyrene shells for our experiments and employed a Baratron pressure gauge and our XRF system to measure the argon content of the shells.

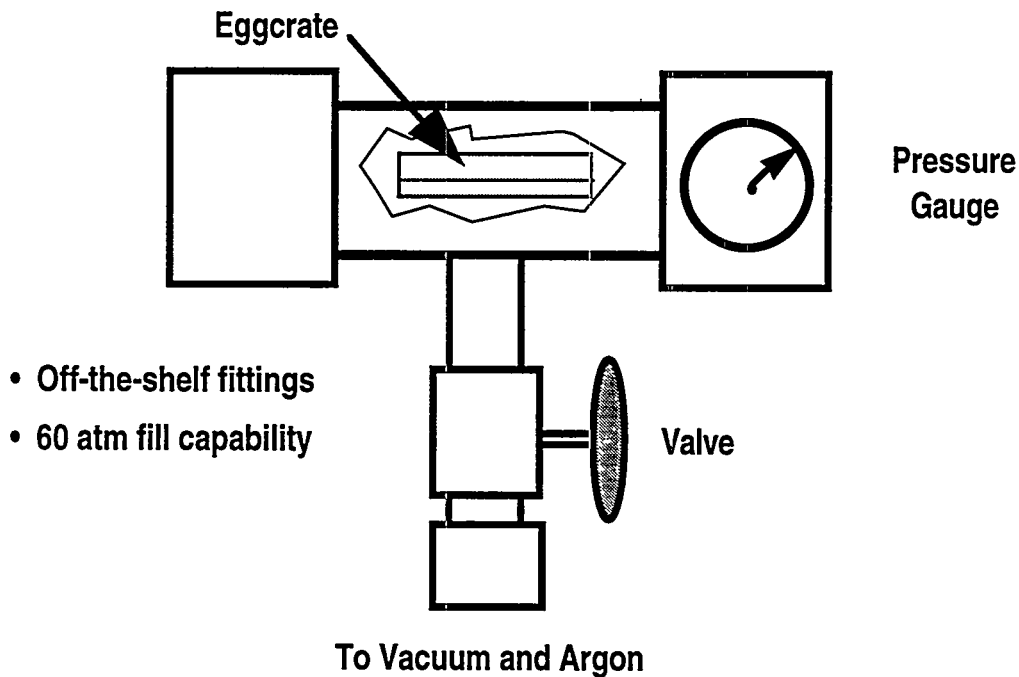


Fig. 2-18. Argon permeation apparatus.

In each experimental run, we loaded fully characterized composite capsules into an aluminum "eggcrate" then loaded the eggcrate into our permeation apparatus. We connected the apparatus to a switchable vacuum/argon gas line. We evacuated the apparatus then backfilled it typically with 500 torr of argon. After sealing the apparatus, we placed it in an oven heated to the desired run temperature. We left the apparatus in the heated oven for a time we deemed sufficient for several hundred torr of argon to permeate into the shells. After removing the apparatus from the oven and letting it cool down, we unloaded the shells and measured their argon content using our XRF system. From the resulting data, we calculated the "half-life" of the argon fill, defined as the time required to fill the capsules to half the pressure of the gas contained inside the permeation apparatus.

We ran this experiment over the temperature range of 102°C to 142°C. The results are shown in Fig. 2-19.

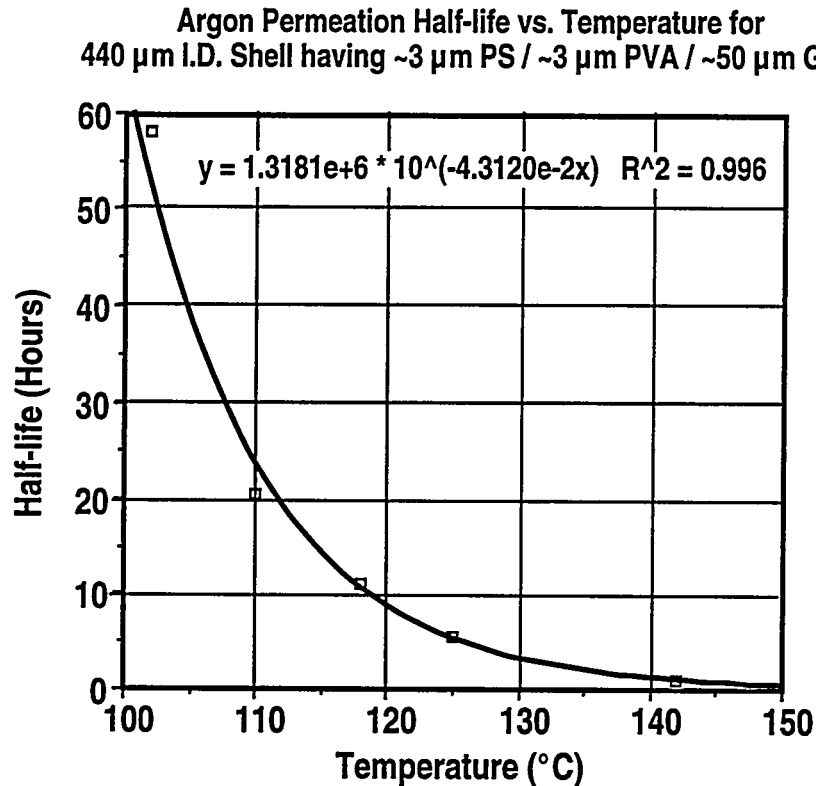


Fig. 2-19. Argon permeation half-life versus temperature for standard composite polymer capsules.

2.4.3. EXPERIMENTAL RESULTS

Our permeation experiments indicated that composite GDP/PVA/PS capsules with an inner diameter of $\sim 440 \mu\text{m}$ and a $3 \mu\text{m}$ thick PVA coating have a fill half-life of approximately one hour at 142°C . Based on this finding, we concluded that capsules will readily fill to within 1% of a desired pressure with an overnight fill consisting of about 20 half-lives.

We were concerned that the inner polystyrene mandrel would deform during our fill procedure. To assuage this concern, we measured the wall uniformity of shells before and after permeation filling. In all cases, we could detect no measurable wall uniformity degradation due to the overnight fill. Likewise, characterization of inner wall surfaces before and after filling revealed no change to the inner surface finish.

To independently confirm that trial capsules had reached desired fill pressures, we broke an assortment of capsules in index matching fluid to measure the size of the resulting argon bubbles. Surprisingly, we found that the gas bubbles released from control capsules, filled with argon using the previous fill technique, were far too large. Capsules filled with 0.1 atm of argon (and confirmed to have 0.1 atm of argon via XRF) had a 0.3 atm sized bubble. On the other hand, composite capsules permeation-filled overnight at 142°C had a bubble size that matched the targeted argon pressure as confirmed by XRF. The most likely cause of the discrepancy between gas bubble size and argon pressure in those capsules filled using the original gas filling technique is air and/or water vapor permeating into the capsule prior to complete drying of the PVA overcoat. Subsequent bubble measurement experiments with bare polystyrene shells which show a half-life for air permeation on the order of a few minutes at room temperature are consistent with this interpretation.

Perhaps the most significant advance made possible by our diagnostic gas fill procedure is a shortened target delivery time to the national ICF laboratories. This attribute results from our ability to now stockpile PVA-coated shells that do not contain argon. When an experimentalist specifies a total wall thickness and argon fill pressure for the capsules, we simply GDP-coat the shells to specification then fill the shells with argon overnight by permeation. Using this procedure, we have reduced capsule delivery time by a factor of two, increased the diagnostic gas fill pressure range (we have delivered capsules filled with 10 atm of diagnostic gases), removed entrapped air from capsules, and improved the accuracy and precision of diagnostic gas fills.

For further information, please contact D. Steinman (GA).

2.5. REFERENCES

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- [2-2] H.K. Lintz, S.R. Murrel, R.L. Crawley and J.C. Daukas, "The Deposition Rate Dependence of Plasma Polymerization on the Radius of Curvature of the Substrate," J. Vac. Sci. Technol. A 6(3), 1869 (May/June 1988).
- [2-3] GA memo ICFT94/225.
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SECTION 3

HOHLRAUM PRODUCTION

3. HOHLRAUM PRODUCTION

We achieved full production capability for the micromachining, dimensional characterization and gold plating of hohlraums this year.

3.1. MANDRELS

This year we achieved full production capability for the fabrication of hohlraums. This includes both the capability for micromachining of copper mandrels and for gold plating of these mandrels. In all, we delivered 726 gold-plated mandrels of 27 different types to LLNL and 48 gold-plated mandrels of two different types to LANL this year.

During the year a high speed milling attachment that utilizes a diamond cutting tool was added to the Precitech Optimum 2000 lathe that we acquired last year. This attachment allowed us to cut flats on mandrels without removing the mandrel from the collet in which it was held for the initial machining. Previously, to cut a flat, the machined mandrel was removed from the collet; a fly-cutting tool was inserted into the spindle of the lathe; the mandrel was inserted in a fixture; and the flat was cut in a fly cutting operation. This required careful positioning of the mandrel and determination of the position of mandrel each time a flat was cut, operations that were very time consuming. With the milling attachment, the mandrel remains in the collet and its position is known. Thus the flat-cutting operation is much faster than was possible previously. Also, the surface finish obtained with the milling cutter is significantly better than that obtained by fly-cutting.

Since there is a detector that measures the rotational position of the spindle on the lathe, the new setup allows us to cut flats at accurately defined positions around a mandrel. An example of this is shown in Fig. 3-1, where 5 flats running the whole length of the mandrel have been cut at equally spaced positions around the mandrel. Mandrels of the type are routinely being fabricated and delivered to LLNL.

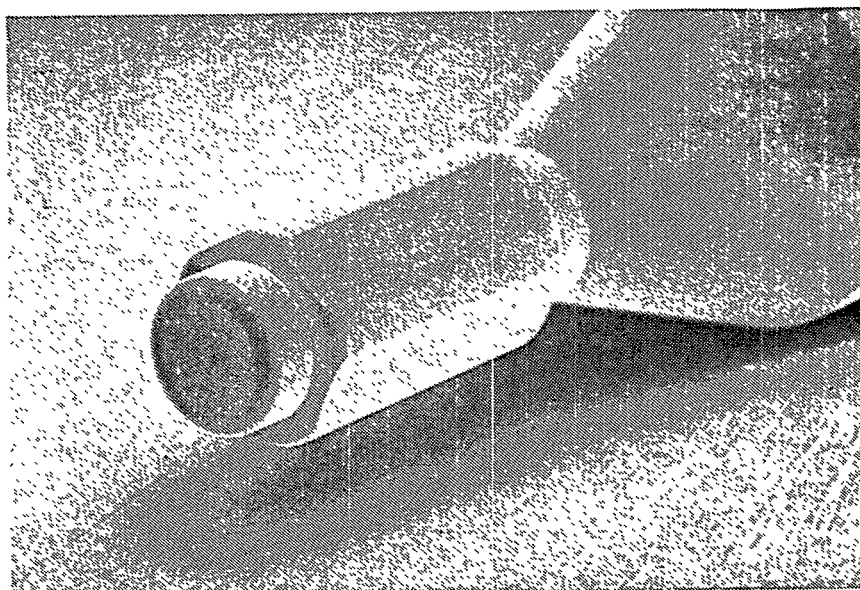


Fig. 3-1. Typical mandrel which has five flats cut at equally spaced positions.

This year we also purchased a second Precitech Optimum 2000 lathe with General Atomics private funds. This lathe is being used to machine ICF components on a part-time basis and has greatly added to our capability. The first Precitech, which was purchased with DOE funds, is set up strictly for production of mandrels and is currently operating 10 hours per day. The setup of the second lathe is changed as necessary either for mandrel production, for production of other ICF components, or for machining tasks on other contracts.

3.2. COMPONENTS

Micromachined components other than hohlraum mandrels include witness plates, Rayleigh-Taylor samples, composite (metals and plastics) mix packages, and miscellaneous one-of-a-kind or unique physics packages. Micromachining equipment, including precision single point diamond turning lathes and characterization interference microscopes, were transferred from the micromachining facility of the DOE Rocky Flats Plant in Colorado to the GA/WJSA facilities. The two phase equipment removal process was begun in FY94 and will be completed in FY95. The first-phase transferred the Rocky Flats #3 diamond turning lathe which was installed for the production of micromachined target components at the WJSA facility in February, 1994. Shortly after installation of the #3 lathe, continuous operation was plagued by a succession of equipment failures,

including spindle blockhead air bearing seizures, air bearing slide failures, and electronic controller malfunctions. While over the course of the year these problems were either resolved or diminished, we managed to develop capability to machine and characterize witness plates and demonstrated machined surface finishes in aluminum of $<100 \text{ \AA}$.

The first development toward witness plate validation involved preparation of aluminum witness plates. Polycarbonate and Crystal Bond #509 potting compound were explored as substrate materials for subsequent aluminum sputter coated parts (performed at LLNL) which are eventually machined into the required witness plates. The Crystal Bond material was investigated because of its ease of casting and its solubility in acetone or MEK. These advantages make the recovery of machined witness plates easier than with the use of polycarbonate, which has been used previously by other machinists. As it turns out, the Crystal Bond has a drawback in that there is an abrasive chemical interaction when diamond turning this material leading to degraded diamond tools. Use of Crystal Bond has been abandoned as a substrate material in favor of polycarbonate. Prior to completion of validating aluminum witness plate fabrication, priorities were changed, and we were asked to produce plastic witness plates either of polycarbonate or polystyrene material. Progress was impeded somewhat by equipment failures with the #3 lathe, but a first iteration of plastic witness plates was generated from thermally-pressed polycarbonate or cast polystyrene solution in aluminum molds which have the desired witness plate profile machined in them. The results of the first trials did not meet specifications. We continued to explore several alternative methods for fabricating plastic witness plates, which are depicted in Fig. 3-2.

In addition to the slump forming of polycarbonate and solution casting of polystyrene into machined aluminum molds, we have investigated direct machining into a cast or thermally-pressed polymer and direct machining into a thin ($\sim 250 \text{ \mu m}$ thick) polymer film. The machining parameters for plastic are different from those for metals, and techniques are being developed to optimize the process. Efforts in producing plastic and metal witness plates will continue into next year.

We were also tasked to machine Rayleigh-Taylor mold samples and begin development of ideas to fabricate a high tolerance, high aspect, tapered-hole physics

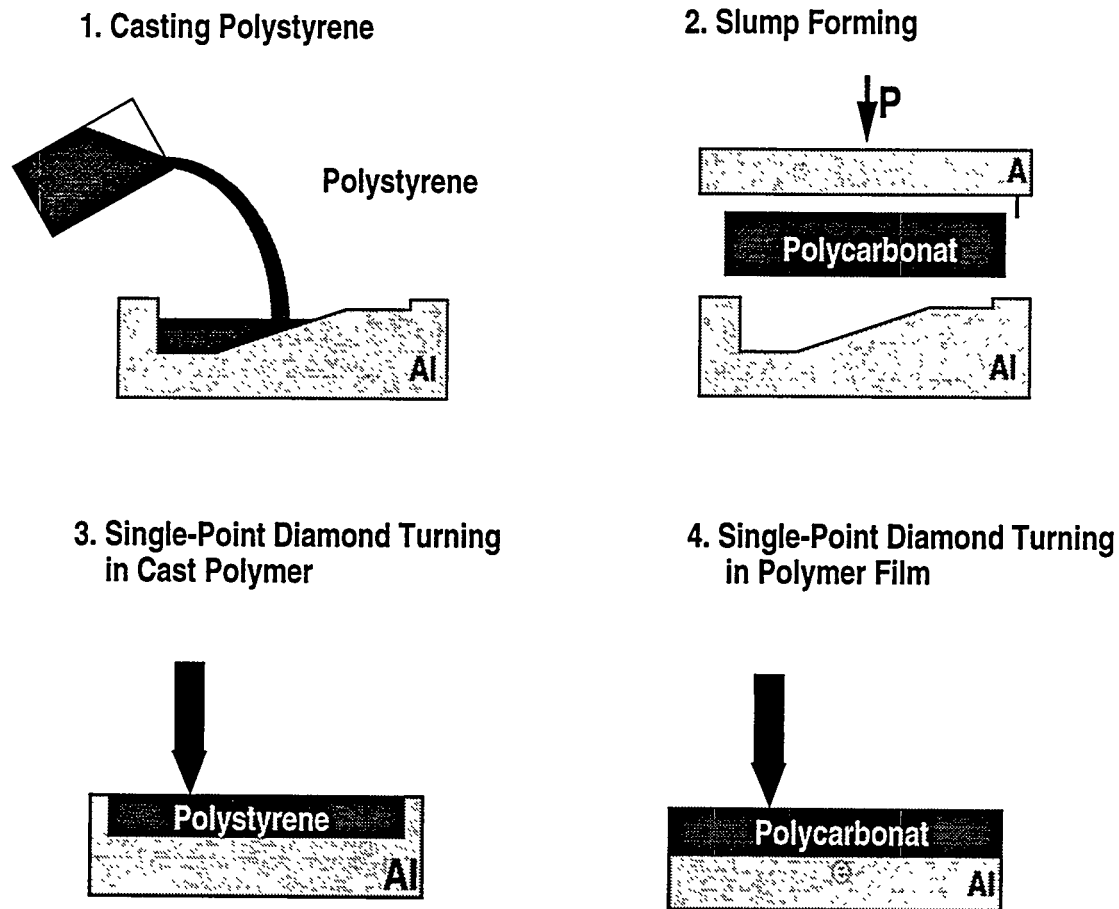


Fig. 3-2. Plastic witness plate development.

package made in plastic. We prepared the machining programs required for the Rayleigh-Taylor samples and actually machined some simple sine wave patterns into aluminum ($\lambda = 200 \mu\text{m}$, $A = 30 \mu\text{m}$) to validate the programs. However, the fabrication of the Rayleigh-Taylor molds has been postponed to FY95.

SECTION 4

OTHER TARGET DEVELOPMENT

4. OTHER TARGET DEVELOPMENT

During FY94 we were tasked to develop the necessary techniques and methods for producing and characterizing NIKE targets and target parts and delivering them to NRL. In addition we fabricated, characterized and delivered foils, filters and polymers to SNL and perdeuteropoly(trimethylsilylstyrene) to LLE.

4.1. NIKE TARGET DEVELOPMENT AND DELIVERY

One of our major tasks during FY94 has been the development of techniques for the production, characterization, and delivery of CH foil targets for the NIKE laser program at the Naval Research Laboratory. Although the targets are predominantly planar films of CH polymeric materials, the specifications for surface smoothness, surface geometry; thickness uniformity, and film flatness provide many challenges for materials preparation and characterization and for the assembly and characterization of the targets.

4.1.1. CH FILM PRODUCTION AND CHARACTERIZATION

The production of CH films with thicknesses in the range 20–200 microns has been a serious challenge this year. We began the year with a process in which a solution of polystyrene was floated on the surface of water contained in a glass beaker [Ref. 4-1]. The films produced by this process usually met our criterion for surface smoothness (surface roughness less than 10 nm) but the film curvature requirements (better than 4 $\mu\text{m}/\text{mm}$) were very difficult to achieve. To solve this problem and still achieve a smooth surface, we decided to cast the films on polished silicon wafers which have a surface roughness less than 1 nm. The surfaces of the films have been characterized by Photon Tunneling Microscopy (PTM) [Ref. 4-2]. The polystyrene films obtained have surface roughness of about 1 nm, which is at least an order of magnitude smoother than required for the NIKE targets. The PTM results have been verified by means of a Zygo scanning interferometer and by a Tencor profilometer. The PTM can provide us with quantitative surface roughness information to the 1 nanometer level.

The surfaces of the polystyrene films cast on silicon wafers are smooth to much better than the required 10 nm but the films as removed from the wafer are very slightly curved. To alleviate the film curvature problem, the film is first heated to 50°C for two hours after being removed from the wafer in order to reduce the overall residual solvent density. The film is then heated to 80°C (the glass transition temperature of the polystyrene film is about 80°C) for one hour to relieve any residual stresses in the film. It should be noted that the films are attached to a metal ring during the drying and thermal annealing processes. The thermal processing enables us to produce targets with film curvatures less than 1 $\mu\text{m}/\text{mm}$.

The curvature of the target films is measured quantitatively by means of a simple technique suggested by John Sethian of NRL. A well collimated beam of light from a HeNe laser, about 2 mm diameter, is incident on the surface of the film to be measured. The orientation of the film is arranged to reflect the incident beam to a convenient screen (a large, white card) located about 75 cm from the film reflection point. If the film is perfectly flat, the reflected spot on the viewing screen will be larger than the incident spot on the film only because of the beam divergence. If the film is curved and deliberately arranged so the beam is incident on the convex surface, the reflected spot on the screen will be larger because of the curvature of the film. It is relatively easy to calculate the film curvature from the values of the film-to-screen distance, the incident laser beam divergence and size at the film and the size of the reflected spot on the screen. Also, the system is calibrated by replacing the film to be measured by a series of reflecting surfaces of known curvature such as the surfaces of a long focal length convex lenses. The curvatures of the surfaces of every target film are measured and provided to NRL together with film thickness, surface quality, composition, volume uniformity and any other parameters significantly related to the target.

Film thickness is measured by accurately locating the two surfaces of the film by interferometric techniques and by using a Sylvac probe accurate to 0.1 μm to measure the distance between the front and rear surfaces of the film. The measurement is accomplished by placing the film in the field of view of the interference microscope and locating the black reference fringe on one surface of the film. The Sylvac probe is set to the position of the movable stage of the microscope and the numbers on the digital readout are noted. The stage is then relocated until the black reference fringe is located on the opposite surface of the film and the reading of the Sylvac is again noted. The difference in the two probe readings gives the thickness of the film to 0.1 μm .

4.1.2. SINGLE AND DOUBLE FOIL TARGET ASSEMBLY

Targets which have been fabricated for NRL during the FY94 year have included single polystyrene films mounted on the small, injection molded, polycarbonate mount [Ref. 4-3]; single unmounted, fully characterized polystyrene films; and prototype targets of several kinds including double foil (polystyrene films), chlorine-doped polystyrene, single foil, rippled, and randomly rough targets.

Targets composed of single films of polystyrene on a polycarbonate mount require very careful assembly techniques but are relatively straight forward to produce. The films are attached to the mounts by an optical grade epoxy cement which is cured very rapidly by exposure to intense ultraviolet light. Other than the use of good mechanical manipulative techniques and the use of appropriately high quality films, the most important factor which determines overall target quality is the geometry of the attachment faces of the target mount. If the surfaces to which the target films are attached are not coplanar, the films will be distorted when they are attached to the surfaces. While we have assembled many targets such that the films are supported on both ends, if the films are sufficiently rigid, we can attach them by only one end. The upper part of the target mount is removed and the film mounted to only the lower "jaw" of the mount. This technique has several positive aspects. First, the film can not be distorted by attachment to two non-coplanar surfaces, and, second, there is better diagnostic access to the film during the laser irradiation of the target. Film mounting by only one end can be done only for films which are sufficiently rigid that bending because of the weight of the film does not occur.

A double film target consist of two films, one 15–20 μm thick parallel to and spaced from another film which may be from 20 to 90 μm thick. The spacing between the films may be between 50 and 300 μm . The two films are to be parallel to 0.5% or better. To assemble the target, the "second" film (relative to the laser irradiation side and 15–20 μm thick) is attached by its two ends to the two coplanar faces of the target mount. Then, two short pieces of film of appropriate thickness are cemented to the mounted ends of the thin film to act as spacers. The "first" film is then cemented to the spacers to complete the double film target. Each of the films used for the target is fully characterized before the assembly. After the films are in place, the curvature of the films is measured and a measurement is made of the parallelism of the two target films.

4.1.3. RANDOMLY ROUGHENED FILMS

Some of the experiments to be done at NRL require targets with surfaces which are randomly rough. Several techniques have been used to produce targets which have surfaces which have a wide spectrum of spatial frequencies. These techniques include: Bumpy mode parylene deposition; electrohydrodynamic spraying of solutions of polystyrene and/or other polymers; and replication of solids known to have broad spectrum rough surfaces.

Parylene can be deposited in a mode which favors homogeneous polymerization of the monomer vapors in the vicinity of the deposition substrate. Also, by adjusting the rate at which the parylene is deposited, it is possible to enhance the generation of irregularities in the deposited material. Thus, randomly rough parylene film can be deposited as a result of some particulate deposition and also from the nucleation of some irregular growth sites on the surface of the depositing parylene leading to "bumpy" surfaces. Such parylene films can be used to fabricate rough targets for the NIKE experimental program.

When a solution of a polymer in a high vapor pressure solvent is allowed to flow at a well controlled rate through an electrically conducting capillary tube maintained at a suitable potential, the liquid is caused to spray from the tip of the capillary in a stream of very small liquid particles. The size of the particles can be controlled to be as small as single molecular ions and as large as several millimeters [Ref. 4-4]. The charged particles produced by these means can be deposited onto substrates to form layers of polystyrene whose surfaces are suitable rough. By controlling the electric field at the tip of the conducting capillary, the fluid flow rate, and other such parameters as the overall local geometry around the substrate, the size distribution of the particles can be adjusted to produce the surfaces required for the NIKE experiments. While some surfaces have been produced by this technique, further development is required to produce targets in large enough numbers to be useful to the program.

Rough surfaced targets have also been generated by the replication of rough solid surfaces by allowing the solvent in a solution of polystyrene to evaporate after depositing the solution on the rough surface. This is a technique which has been used for many years in the field of electron microscopy to produce transmission electron microscopy targets from samples which are opaque to the electron beams. The polymer reproduces faithfully the details of the surface on which it is cast. We have produced rough targets by

replicating the rough surface on the back of a silicon wafer which has a relatively wide spectral distribution of spatial wavelengths in its surface.

4.1.4. RIPPLED TARGETS

Some of the experiments to be done at NRL require targets whose faces may be generally described as having $\sin(x)\sin(y)$ ripples on the surface. Thus, the surface of the target film may be described by the height, $z(x,y)$, of the surface above some arbitrary x-y reference plane in the target film. The function may be simply a series of sinusoidal waves on the surface

$$z(x,y) = z_0 \sin(kx)$$

where z_0 is the amplitude of the waves, $k = 2\pi/\lambda$ is the wave number and λ is the wavelength of the waves on the surface or may be more complex surface patterns.

A method we have used to produce prototype targets with rippled surfaces is the replication of a metal surface which has waves machined into the surface with a single point diamond tool. The polymer replica of the surface is a faithful reproduction of the machined surface and a virtually unlimited number of castings can be made from the same surface. Other techniques which have been examined but which require further study include replication of silicon wafer surfaces etched to suitable patterns and casting of films on the surface of dense liquids in a ripple tank.

For further information , please contact Dr. C. Hendrix (WJSA).

4.2. FABRICATION OF FOILS, FILTERS AND POLYMERS

The Foils, Filters and Polymers Task, which was carried out for Sandia National Laboratory (SNL) was divided into five subtasks. The five subtasks are discussed separately below.

4.2.1. FABRICATION OF FOILS

The foils fabricated in this subtask consisted of freestanding aluminum foils supported by either 0.5 in. or 1.0 in. diameter circular frames, freestanding gold foils with fiducial holes supported on oval titanium frames 1.5 in. by 1.5 in. in dimensions, and

duplex gold-aluminum foils on similar oval frames. Altogether, 321 of these foils were fabricated and delivered to SNL.

The metal foils were deposited by sputtering on a cellulose acetate film which was subsequently dissolved to leave the metal foil free standing. The detailed fabrication sequence consisted of: (1) cement the cellulose acetate film to a large carrier frame; (2) deposit the desired metal by sputtering on the cellulose acetate film; (3) cement the desired frame to the metal side of the composite film; (4) cut around the frame to remove the frame from composite film attached to the large carrier frame; (5) dissolve away the cellulose acetate film using amyl acetate; (6) punch fiducial holes in the free-standing foil if desired.

Originally, we had used acetyl acetate to dissolve the cellulose acetate film, but this year we switched to amyl acetate because we found that the latter solvent induced less stress in the foil during the dissolving process. Finally, the thickness of each foil was characterized by alpha particle spectrometry at several positions before delivery to SNL.

A large box coater was received as part of the equipment available from KMS. This coater was installed in our laboratory and was made operational under the work carried out in this subtask. We plan to use this coater to deposit foils in FY95.

4.2.2. FABRICATION OF FILTERS

The filters fabricated in this subtask consisted, principally, of aluminum foils on Mylar films although a few free-standing boron nitride filters were also fabricated. Altogether, 105 filters were delivered to SNL this year. Most of these foils were sandwiches made up of 0.2 μm of aluminum, 1.5 μm of Mylar and 0.2 μm of aluminum. The aluminum was deposited by sputtering, and the filters were mounted on circular frames either 2 in. or 2.5 in. in diameter.

As with the foils, the thickness of each filter was characterized using alpha particle spectrometry before delivery to SNL. For the sandwich filters, this involved characterizing the thickness of each component of the filter.

An ion plater obtained from General Atomics surplus equipment was installed at WJSA to give us increased capability for deposition of elements and compounds. Thus,

we now have capability for deposition of films with sputtering, evaporation and reactive evaporation processes.

4.2.3. SUPPORT TO SNL

An engineer, Tom Alberts, is assigned to Sandia National Laboratory in Albuquerque, New Mexico. His major tasks are to act as a liaison between GA and SNL, fabricate foils and filters with SNL facilities, and develop x-ray characterization of filters. Tom worked with SNL personnel to fabricate a large range of foil designs for use as x-ray filters in the SNL 1994 Target Series. An improvement in the quality of foils through a reduction in the number of pinholes in the foils was obtained when the glass slides previously used as substrates for deposition were replaced with silicon wafers. This occurred because the silicon wafers were received in a very clean state. Rigorous cleaning of glass slides did not achieve the same cleanliness as was present in the as-received silicon wafers.

The free-standing filter designs fabricated at SNL for the 1994 Target Series are listed in Table 4-1.

The design of the new EST diagnostic required filter materials to be coated directly onto a scintillator material. Tom applied the scintillator to a fiber optic face-plate using spin coating techniques developed by SNL personnel. He also developed processes to coat this scintillator with films of 0.2 μm Al, 0.35 μm Al/0.8 μm Ti, 0.6 μm Cu, 0.6 μm Fe, 0.3 μm Ti, and 1.0 μm Ti. These processes included techniques to minimize stress, peeling, and cracking of the films.

Tom rebuilt an x-ray attenuation measurement system from existing equipment. The system incorporates a single anode Mason system capable of generating x-rays of energies below 5000 eV. This system became fully operational at the end of FY94. It will be used to measure selected filters used in the 1994 Target Series and filters to be used in the 1995 Target Series. These measurements will be compared to measurements taken with alpha spectrometry in order to generate a table to cross calibrate data between the two methods of measurement.

Table 4-1
Filter Designs Fabricated at SNL for the 1994 Target Series

Single Layer	Double Layer	Triple Layer
0.2 μm Al	0.11 μm Al/0.66 μm Ti	2.0 μm Pary-D/0.4 μm Al/0.2 μm Sn
0.35 μm Al	20 μm Pary-D / 50 μm Al	0.2 μm Al/0.3 μm Sn/0.5 μm Ti
1.0 μm Ti	7.0 μm Al/2.5 μm Cu	2.5 μm Pary-D/0.5 μm Al/0.3 μm Sn
0.7 μm Zn	1.0 μm Cu/6.0 μm Mylar	3.0 μm Pary-N/0.2 μm Al/0.2 μm Sn
1.2 μm Pary	0.4 μm Cr/6.0 μm Mylar	3.5 μm Pary-N/0.2 μm Al/0.3 μm Sn
	2.7 μm Pary-D/0.2 μm Sn	
	26 μm Pary-D/0.3 μm Al	
	0.3 μm Pary-N/0.5 μm Al	
	2.5 μm Pary-N/0.3 μm Ti	
	3.5 μm Pary-N/0.2 μm Sn	

4.2.4. DEUTERATED PARYLENE

The deuteration of parylene (2.2-paracyclophane, $\text{C}_{16}\text{H}_{16}$) was attempted by several routes. In all cases, the deuteration methods are well known in the literature, and we simply sought to apply them directly to the parylene molecule. We felt that trying to deuterate parylene, where the carbon backbone was already formed, was preferable to deuterating a precursor, and then trying to make parylene in 10%–20% yield at best. Levels of deuteration were determined by comparing the gas chromatography-mass spectroscopy (GC-MS) spectra of the deuterated parylene to the non-deuterated parylene.

The initial attempts involved the direct exchange of H for D. In one method, we put parylene, platinum black, and deuterated benzene (C_6D_6) in a Parr pressure bomb, and heated the mixture to 130°C–180°C. In this case, the benzene is both the source of deuterium, and the solvent for the reaction. We obtained a material near fully exchanged, but its mass (on the mass spectrometer) was four units higher than the completely deuterated $\text{C}_{16}\text{D}_{16}$. Apparently, two extra D atoms have been incorporated into the molecule. We think the parylene side chain separates on the Pt surface, leaving two

radical ends. Two D atoms on the Pt surface then react, leaving a terminal $-CD_3$ group. The two ends then cannot recombine to form parylene. This approach was abandoned. A second attempt involved stirring D_2SO_4 with parylene, or with parylene plus deuterated benzene. In both cases, no deuteration was observed over multiple weeks. Apparently, the parylene solubility is so low, that bulk reaction did not occur.

In a third attempt, the exchange was attempted by removing a proton with butyl lithium (BuLi), to produce an intermediate lithiated material. This lithiated parylene is then exposed to D_2O , to replace the Li with D (and form LiOD, which is washed away). However, in several solvents, no lithiation occurred. Parylene is apparently one of very few molecules which is not metallated (by lithium) under these severe conditions.

Because the direct exchange of H for D was unsuccessful, we then attempted the synthesis of brominated parylene, followed by the reaction of BuLi and D_2O , to put a D where the Br atoms were located. Bromination of the parylene rings was done with $FeBr_3$ and Br_2 in CH_2Cl_2 , in a 22 l flask, to produce some 300 g of tetrabrominated parylene. The GC-MS showed the material has 2 bromines on each ring, as three isomers. Addition of this mixture of brominated isomers to BuLi, followed by addition of D_2O , produced a mixture of the brominated isomers and pure tetradeuterated material. This mixture was never separated. To circumvent this mixture, the brominated mixture was added to ether, and the ether cannulated into a BuLi solution. The ether was distilled back to the starting brominated parylene, to pick up more material, and added again to the BuLi. This cycle was repeated multiple times, with each cycle taking about 1 hour. The process produced pure tetralithiated material, but at a very low rate (less than 1 gram/hour). Hydrolysis with D_2O gave pure tetradeuterated material. If the lithiation cycle was continued for more than 12–24 hours, one could observe the growth of a butyl substituted parylene material. Thus, the lithiation cycle could only be done for 12–18 hours, after which the hydrolysis was done. Typically, upon hydrolysis and work up of the material to obtain the solid tetradeuterated parylene, some 3–5 grams of pure material was formed after some 18–24 hours of labor. Apparently, the other isomers were not soluble in the ether, and thus the lithiation reaction could not occur. Yields of deuterated material from the same large batch of brominated material declined from one run to the next, apparently exhausting the soluble isomer. Ultimately, from about 150 grams of original parylene, I obtained 11.5 g of parylene- D_4 ($C_{16}H_{12}D_4$), with much residual tetrabrominated parylene. This level of deuteration is ideally 25.0%, and GC-MS analysis of several

batches were typically 22.5% deuteration. (The presence of the butyl-parylene, where the butyl has H, lowers the deuteration level slightly.)

Bromination of the ethyl bridges was done with AIBN, Br₂, and light in CH₂Cl₂, to form a dibrominated material. (Stoichiometrically there was enough Br₂ present to make the tetrabrominated material, but after several days and repeated addition of AIBN, only the dibrominated material was obtained.) Addition of the dibrominated material to BuLi/ether resulted in immediate reaction, to form the dilithiated material. Upon hydrolysis, the dideuterated material was formed, which ideally has 12.5% D. The 14.1 grams delivered had a deuteration level of 10%, again with the butyl-parylene lowering the level slightly.

Ultimately, this route of deuteration via bromination is very slow, most likely due to the limited solubility of parylene in virtually all solvents. This very limited solubility leads to significantly reduced yields and is not of practical use in the synthesis of large quantities of deuterated material.

4.2.5. FOIL SPHERES

This subtask was initiated this year to investigate the feasibility of fabricating relatively large-diameter (~5 mm) foil spheres by depositing the foil material on polystyrene shells, drilling a small hole through the foil material on each shell, and then dissolving out the polystyrene.

For the feasibility tests we decided to work with spheres in the size range 3 mm to 5 mm in order to ease the production of the shells. Screening experiments proved that the microencapsulation "shake and toss" technique could produce shells of the desired size, but the production rate of spheres of the desired size was low. A simple droplet generator was made to produce microencapsulated shells which increased the production rate. However, the size distribution was large. More than 300 shells were produced for testing. All of the larger shells were vacuole laden although this was not important since the shells were to be dissolved once they were coated with gold. Occasional small shells were vacuole free. An example of an uncoated shell is shown in Fig. 4-1.

Initially we tried to coat the shells by sputter deposition while the shells were agitated in a pan. The appearance of a shell coated in this way is shown in Fig. 4-2. The

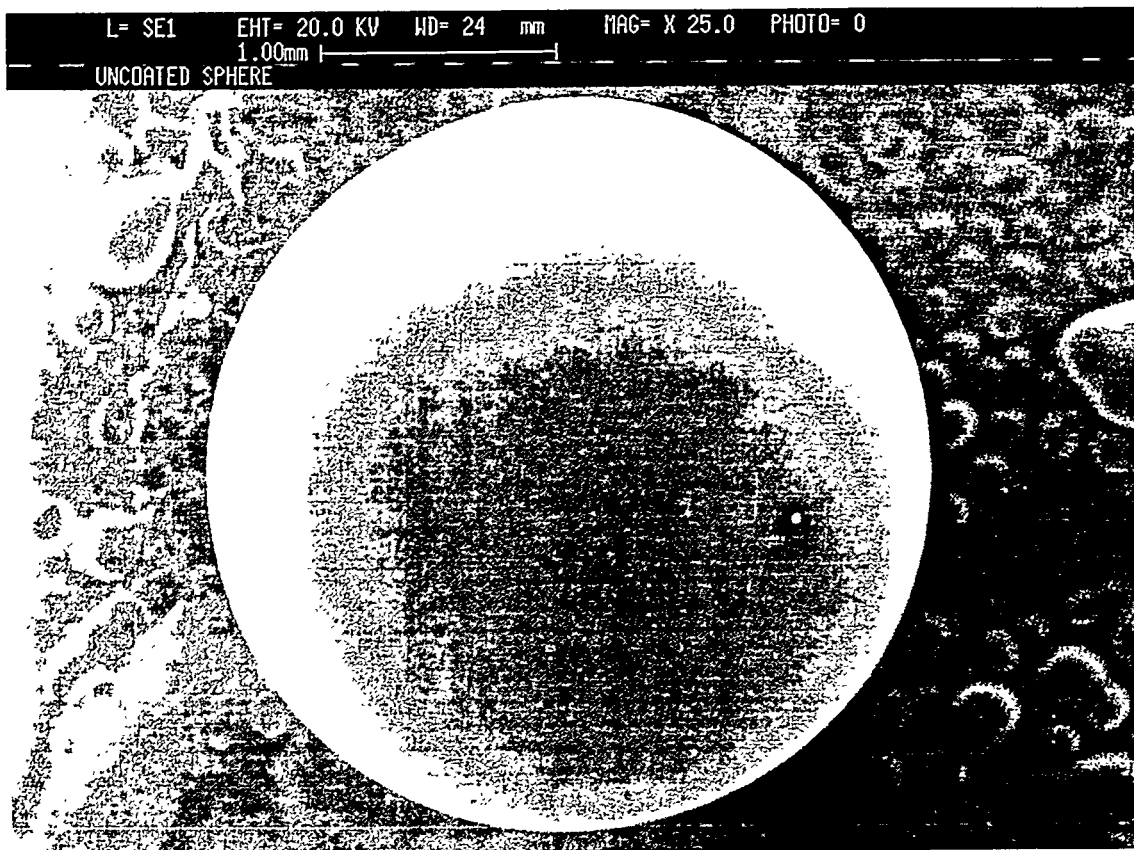


Fig. 4-1. A typical uncoated microencapsulated shell.

gold coating is very rough, apparently due to sticking of the “fresh” gold surface on the sphere to fresh gold on the pan. Thus, it did not appear that sputter deposition of gold on the shells was practical.

A small hole was drilled in the gold coated shell of Fig. 4-2. It can be seen on the top of the shell, and the polystyrene was dissolved out, thus proving the feasibility of this operation.

We then switched to electroplating for gold deposition on the shells. To do this a very thin layer of gold was deposited on shells by sputter deposition to provide a conductive surface. The shells were then transferred to a small mesh drum which was rotated while gold was deposited by electroplating. We discovered that cleanliness was very important if defect-free coatings were to be obtained. An example of a shell coated

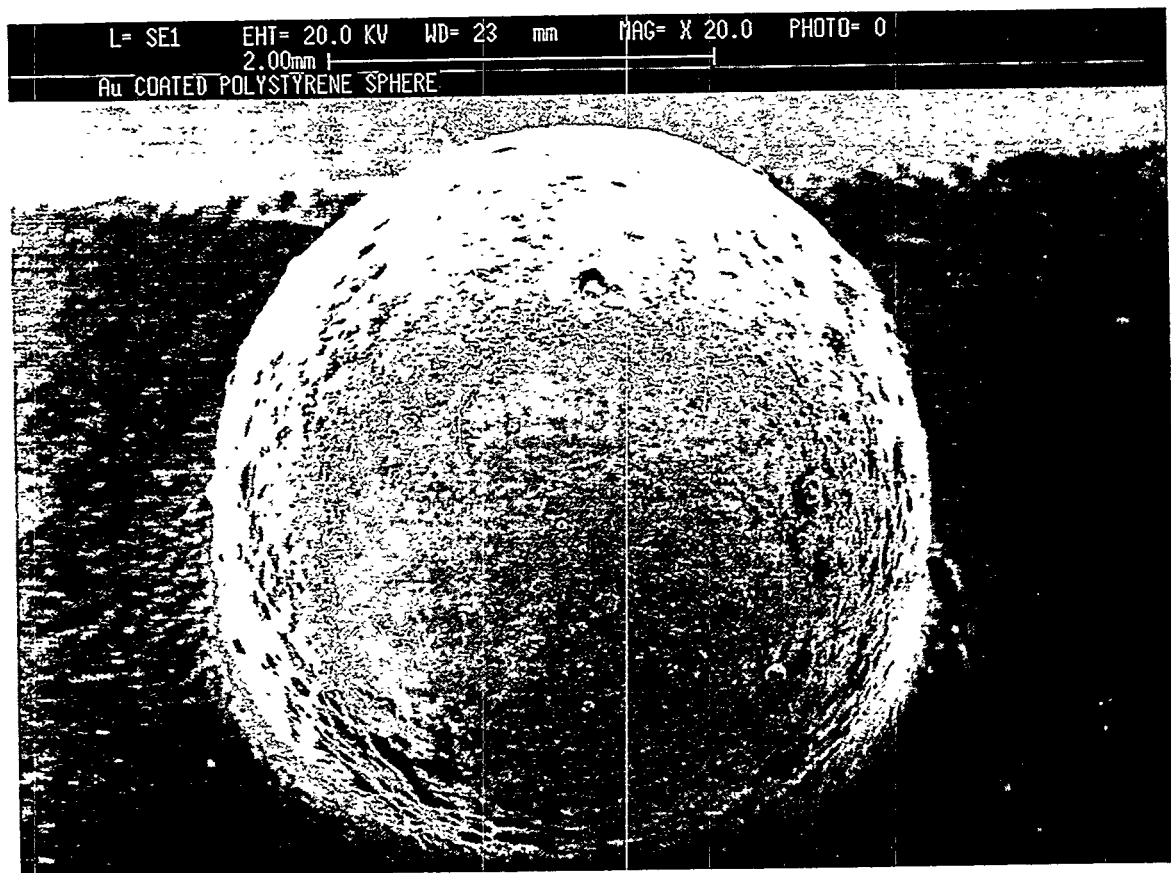


Fig. 4-2. Typical appearance of gold sputter coated shell.

when little attention was paid to cleanliness is shown in Fig. 4-3, and an example of a shell coated when the shell was carefully cleaned before coating is shown in Fig. 4-4. There are still a few defects in the gold layer on the shell shown in Fig. 4-4, but otherwise the coating appears of high quality.

This study demonstrated that electroplating of gold on polystyrene shells followed by dissolution of the polystyrene through a small hole is a practical way of producing gold spherical foils.

For further information, please contact Dr. J. Kaae (GA).

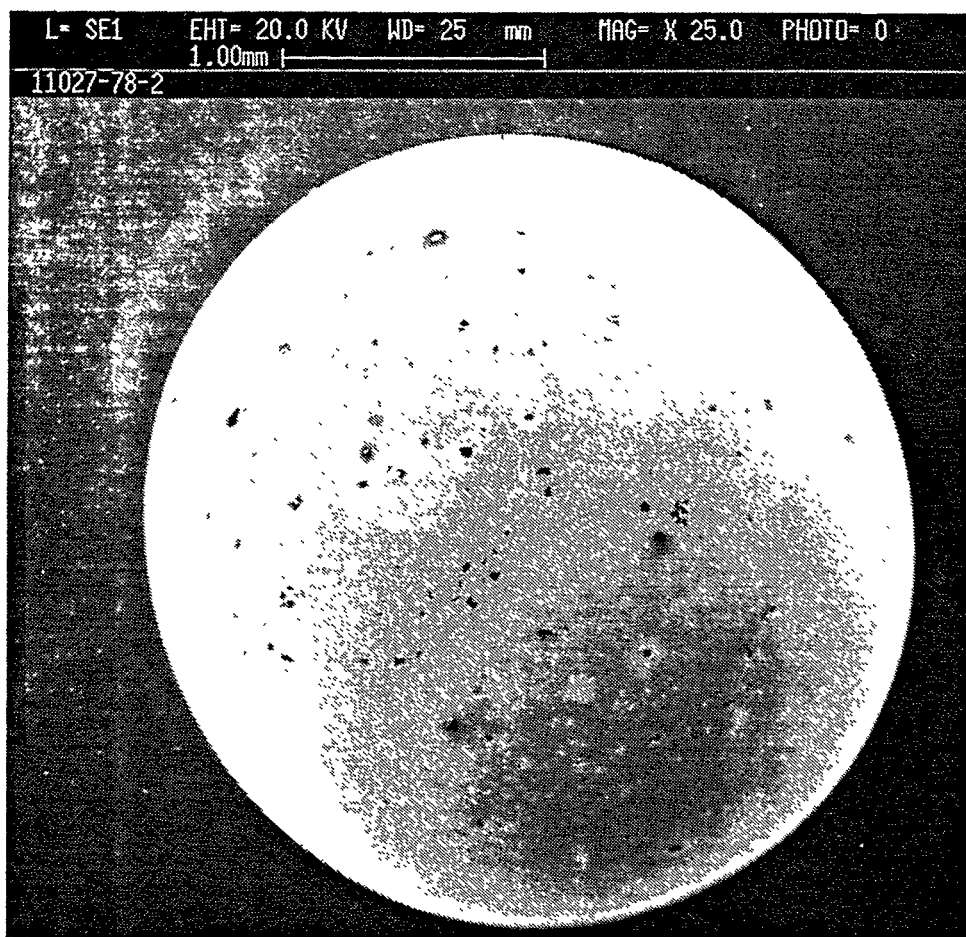


Fig. 4-3. Example of a coated shell with no cleaning.

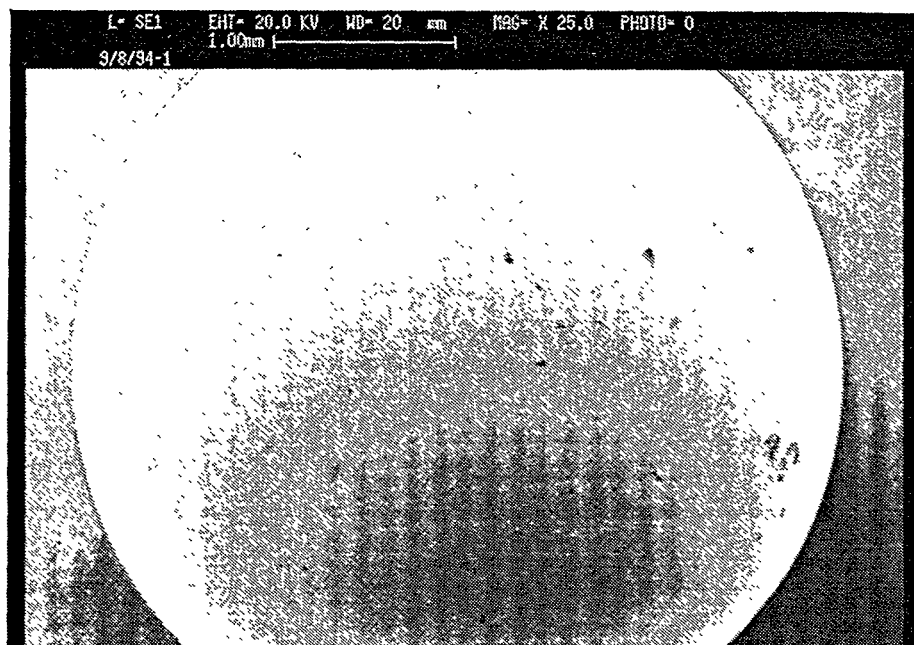


Fig. 4-4. Example of a clean and coated shell.

4.3. CHARACTERIZATION DEVELOPMENT

This task was comprised of three characterization development subtasks.

4.3.1. PHOTO-NUCLEAR REACTIONS MEASUREMENT METHODS

A study has been undertaken to evaluate the potential of gamma-neutron reactions to measure the deuterium content of filled inertial confinement fusion capsules. Because various hydrogen isotopes (hydrogen, deuterium, and tritium) have different diffusion rates, the relative isotope concentrations in a capsule can vary from the relative concentrations in the liquid fill system if the capsule is not filled to equilibrium pressure. A non-destructive method of analyzing the fill concentrations in a capsule could verify that the desired isotope ratios have been achieved and could also be used to detect fuel loss due to capsule leaks.

Gamma rays interact with different elements in ways which produce unique radiation signatures for each element. The (γ, n) reaction was chosen because the threshold energy (the energy below which the reaction will not occur) of 2.23 MeV for this reaction in deuterium is significantly below the threshold energies for this reaction in all other elements except Be. Therefore, as long as the capsule contains no beryllium, a gamma source above 2.23 MeV (and below ~5 MeV) can only produce neutrons by interacting with deuterium. This allows an accurate measure of the deuterium concentration from the neutron production rate and the source strength. Because neutrons deposit energy in a scintillation detector over a larger time interval than gammas do, discrimination between neutron and gamma signals is possible with a combination pulse shape analyzer and timing single channel analyzer.

An experiment to measure deuterium in a filled capsule would consist of a gamma source, a capsule holder, neutron/gamma detectors on either side of the capsule holder and behind the capsule (giving a coverage angle almost equal to 4π), and a multi-channel analyzer (MCA) to sort the detector signals by decay time (and thus separate the neutron counts from the gamma counts).

The accuracy of the deuterium measurement depends on the total number of neutrons created and counted. A measurement containing at least 1000 counts is needed

for accuracys of a few percent. To limit experimental runs to less than a day requires count rates above 40 counts/hour, or 0.012 counts/second. Unfortunately, these count rates require large gamma-ray fluxes, and therefore require strong gamma sources. Particle accelerators can be used as sources of directed gamma rays, but the cost of such devices may require moving the experiment to an existing source.

Although the neutron fluence created in this experiment will be small enough that it will not create any measurable capsule damage, there is a concern that high intensity, high energy, gamma radiation could damage the plastic (or glass) capsule shell. Radiation damage to polystyrene structural properties requires 10^4 to 10^5 Gy (1 Gy = 1 J/kg), so the capsule damage should be negligible [Ref. 4-5].

Conclusions And Recommendations. The fundamental conclusions from this study are as follows:

1. Photo-nuclear reaction measurement (gamma-neutron reactions) using a gamma source with gamma energies above 2.23 MeV and source strength on the order of a Curie could be used to measure the deuterium content of a filled capsule.
2. The photo-nuclear reaction measurement would be non-destructive, so that a capsule could be used after a measurement.
3. The cost of setting up and operating an experimental area using a gamma source with the required strength is prohibitive. This measurement technique will only be practical if affordable access to an existing high-energy gamma source can be arranged.

The results of this study lead to a recommendation not to pursue an experiment at the present time.

4.3.2. COMPUTER ASSISTED SHELL SELECTOR (CASS)

Polystyrene shells are used as the mandrel in current ICF composite plastic capsule designs. About 2% of the polystyrene mandrels after sieving make it through the various coating steps to become finished capsules. The low yield and large number of finished shells needed require a technician to characterize many mandrels for any given delivery. The characterization can be repetitious and time consuming. The CASS was proposed by

Joe Mauger at LLNL as an instrument to help automate initial characterization and selection of polystyrene shells. We completed assembly of a prototype CASS at LLNL this year.

Functional Requirements. The objectives of the CASS system are to:

1. Characterize the diameter and eccentricity of mandrels.
2. Measure wall thickness of the shells.
3. Remove the accepted shells for continuation on the successive steps in the fabrication process.

Equipment and Layout. The CASS system consists of a test stand which includes:

1. A motorized x-y translation stage with a mirror mounted on top. The shells are placed within a 4 inch square area on the mirror.
2. A Leitz-Wetzler microscope equipped with 3.2x, 5x, and 10x objectives. The microscope also has an interferometer accessory that is attached to an eyepiece when we wish to measure shell thickness.
3. A shell picker.
4. A computer data acquisition and control system.

Operational Results. We performed a series of measurements to quantify the operational characteristics of the CASS system. Tests were run to determine precision in the diameter and wall thickness measurements. The system consistently identified shells. After the measurements were completed, the shells were removed from the mirror surface and the diameter and wall thickness manually measured by Charlotte King of LLNL. Table 4-2 contains a sample of our data.

We next performed a series of runs to time our measurements. We found it would take 5 hours to scan a 4x4 inch surface containing 2000 shells assuming half of them were good. Wall thickness measurement takes up to 3 minutes per shell.

Table 4-2
Shell Data by CASS and Differences(Δ) from Manual Measurements by Charlotte King

Shell No.	Diameter	Δ Diameter	Sphericity	MSE	Wall Thickness	Δ Wall Thickness
1	396.7	4.8	0.99	0.16	4.19	-0.44
2	422.2	2.3	1.00	0.17	3.60	-0.07
3	423.8	3.2	0.99	0.33	4.19	-0.44
4	425.8	2.2	0.99	0.06	3.75	0.12
5	428.5	3.0	0.99	0.17	3.52	0.23
6	432.6	2.4	0.99	0.33	3.39	0.37
7	432.7	4.3	0.99	0.34	3.85	0.25
8	434.6	3.4	1.00	0.24	3.54	-0.02
9	440.4	3.1	1.00	0.19	3.45	0.54
10	447.5	2.5	0.99	0.22	3.68	0.19
Average	428.5	3.12	0.99	0.22	3.72	0.07
Std Dev	13.6	0.87	0.00	0.09	0.28	0.32

MSE is a measure of goodness of fit of the shell edge to an ellipse. The other columns are self explanatory. All units are in microns.

Recommendation. The possibility of using CASS as a production tool at General Atomics was considered. GA currently sorts polystyrene drop tower mandrels by sieving. They are capable of rapidly separating shells into 5 micron tolerance batches. The shells do not appear to be marred by the sieving process. Wall thickness tends to be very consistent and non-sphericity does not seem to be a factor. Given these statements, CASS cannot keep up with the shell selection speed of GA's current technique and should not be pursued any further if this were its only possible function.

There are other possible uses for the CASS. Micro-encapsulated shells and glass shells are not as uniform in wall thickness as the polystyrene drop tower shells. The wall thickness measurement capability may be of use in those cases. Presumably, these other types of shells could be sieved for diameter. Another use may be in carrying out screening studies of shell production processes. CASS gives fairly complete

measurements, (diameters, eccentricity, wall thickness) of a large number of shells. The data comes in a convenient format for performing further statistical analysis.

4.3.3. FEASIBILITY OF MICRO-CALORIMETRY AS A MEANS OF DETERMINING ICF CAPSULE FILL

We made a proposal about one and a half years ago to look at an alternative way of measuring fill pressures. We proposed to look for a thermal effect due to the change in heat capacity of the liquid-gas system as it undergoes the phase transition into an all gas system at the dew point. This is a slight twist on Sanchez and Upahdye's [Ref. 4-6] optical method of measuring dew point temperatures. There is a large change in heat capacity at the phase transition and the temperature can be measured by using a calorimetric technique. We have successfully measured the dew point on an 800 μm diameter glass shell filled with 50 atmospheres of D_2 at room temperature. This was made possible by using a very sensitive technique known as AC calorimetry.

We have previously reported on this project [Ref. 4-7]. This report will be limited to new results and recommendations for future improvements.

Results and Data. Our first successful calorimeter experiment had a D_2 filled glass shell (Shell #1). The shell was measured to have a fill of 48 atmospheres by the optical dewpoint machine. This corresponds to a dew point of 29.08 Kelvin. As can be seen in Fig. 4-5, we measured a dew point of 28.75 K. This corresponds to a fill pressure of 44.87 atmospheres, a 6.5% difference. A second shell from the same batch, (Shell #2), had a calorimetrically measured dew point of 28.74 K (Fig. 4-6). It has not yet been measured with the optical dewpoint machine. Ignoring systematic error, our determination of the dewpoint temperature from our graph is accurate to 40 mK. This corresponds to a 1% error in fill pressure.

The triple points were also measured for Shells #1 and #2. Shell #1 had a measured triple point of 18.55 K. Shell #2 had a triple point of 18.45 K. The triple point of D_2 is 18.73 K. All temperatures reported are those measured by the substrate thermometer. These measurements are also precise to within 40 mK. If we attribute the depression of the triple point temperature to H_2 impurities in the D_2 gas, we calculate 3.8% H_2 in Shell #1 and 5.9% H_2 in Shell #2.

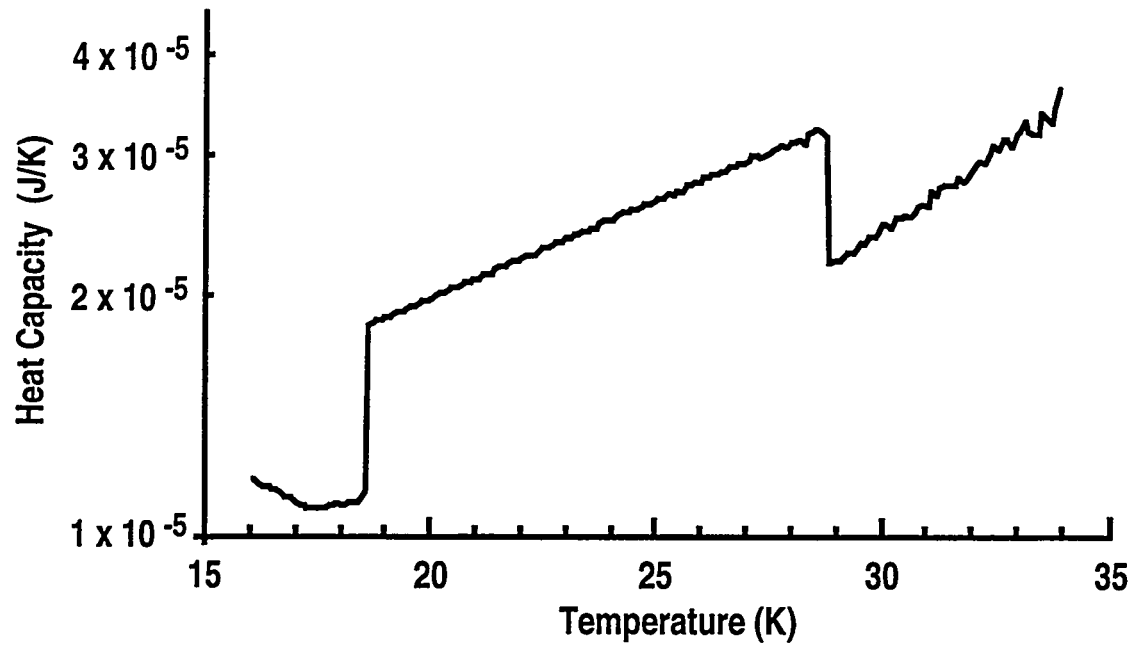


Fig.4-5. Dew point is 28.75K. Triple point is 18.55 K.

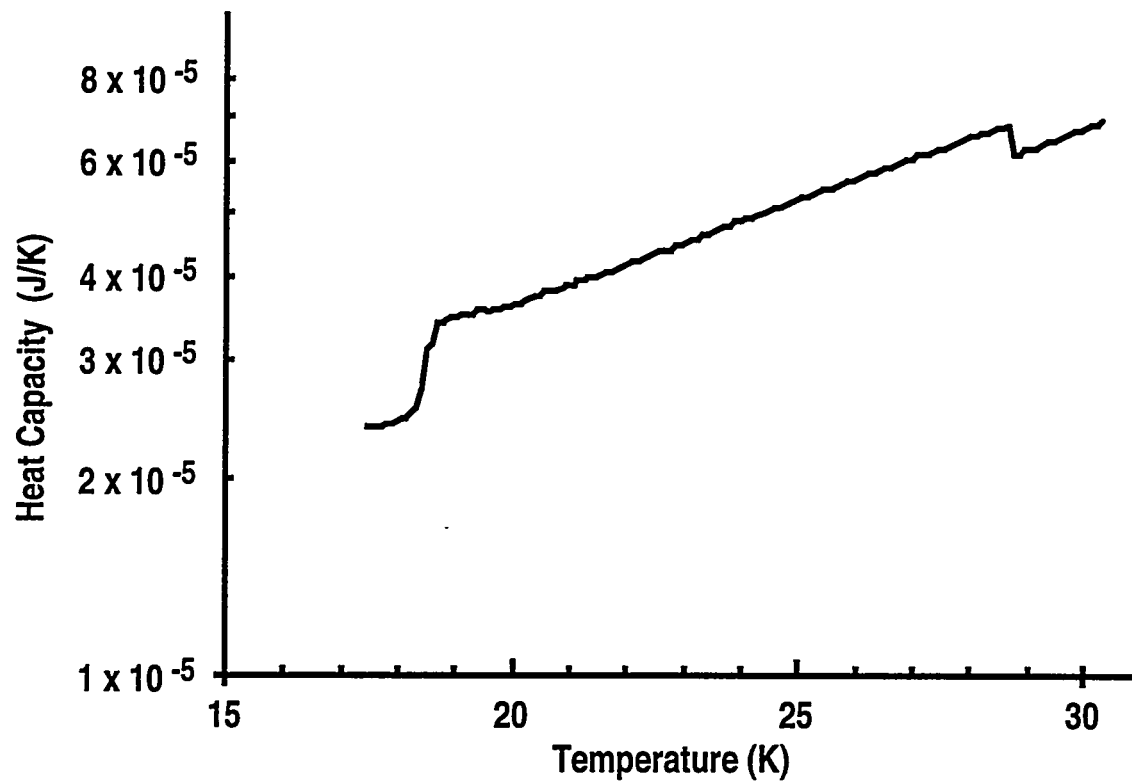


Fig. 4-6. Data using PVA as adhesive. Dew Point 28.74 K. Triple point 18.75 K.

It should be noted that shell #2 was attached to the sapphire substrate with polyvinyl alcohol, a water soluble adhesive. This allows the shell to be removed from the calorimeter without destroying it or the calorimeter.

Recommendation For Use As Production Instrument. We have successfully measured the fill of an 800 micron diameter glass capsule filled with 50 atmospheres of D₂. By scaling based on our signal to noise we have estimated an upper limit for our sensitivity. It should be possible to detect a measured change of 2×10^{-6} J/K in heat capacity. This corresponds to measuring the fill of an 800 micron diameter capsule containing 1.3 atmospheres of D₂. This does not assume any improvement in thermal contact between shell and calorimeter. With future design improvements, we should be able to do even better. It is difficult to estimate the size of the error bars on such a low pressure fill, especially since we have not made such a measurement. The 2×10^{-6} J/K is a detection limit. The accuracy of the fill determination depends on how closely the transition temperature is determined. The best way to determine the error bars, of course, would be with a series of low pressure measurements.

This technique can be used on opaque capsules with no intrinsic loss of sensitivity. The measurement is independent of operator bias. We have demonstrated our ability to attach and remove glass shells from the substrate in a non-destructive manner with PVA (polyvinyl alcohol), a water soluble substance. The thermal conductivity of PVA is adequate for our purposes, and it is easily cleaned from any shell that can be exposed to water.

A final consideration is the suitability of this technique for advanced target designs with very high fill pressures. There is a range of fills near the critical density where this technique will not work. The size of this range remains an open question that should be answered with further work. Once we are far enough beyond the critical density our technique will again work. The difference is we look for the temperature of the transition from a gas-liquid system to an all liquid system. (At lower fill pressures the system transforms from a gas-liquid system into an all gas system.) The only question at this point is will the capsule wall be able to withstand the pressures necessary to make the measurement. The pressures that must be withstood can be found in Souers' book [Ref. 4-8]. In Table 10.6 on Page 139, we find data on a 1.47×10^8 Pa fill. The transition temperature occurs at 34.84 K and the pressure will be 1.02×10^6 Pa. Pressure increases

rapidly on the high temperature side of the phase transition. dP/dT is approximately 0.9×10^6 Pa/K.

We conclude that calorimetric measurement of fuel fill in capsules is a viable technique. It works on current capsule designs having fills of moderate pressure, and given a strong enough capsule, it will work on advanced designs with high pressure fills.

Future Improvements For A Working Instrument. The calorimeter's current design can be improved. The highest priority is to develop a capability to measure multiple shells at one time. The best solution is to attach multiple calorimeters to a single large cooling source. It may be possible to use semi-conductor technology and make multiple calorimeters on a single wafer of silicon [Ref. 4-9].

Another area that needs further development is ruggedization of the assembly. Silicon based calorimeters may prove to be more rugged than our kevlar supported sapphire. The expense of this type of technology should be considered.

Finally, we propose that the production instrument use a mechanical refrigerator. This would be more convenient for operational use and cheaper in the long run than buying liquid helium. A very brief investigation indicated that a mechanical refrigerator necessary for our purposes would cost under \$15K in 1994.

For further information, please contact Dr. J. Sater (WJSA).

4.4. MICROENCAPSULATION OF LARGE SHELLS

The overall goal of this task was to develop the ability to produce large polymer shells via controlled-mass microencapsulation. By attacking the problem using a combined experimental and theoretical approach, we were able to achieve nearly all of the goals set at the beginning of the fiscal year for this challenging task. The systematic approach we pursued involved breaking the problem down into three areas — controlled-mass microencapsulation; sphericity and concentricity of the product shells; and vacuole elimination. In addition, to help us understand the role of processing parameters in achieving concentricity, finite element mass transfer modeling was of particular importance. Although experimentally we occasionally have been successful in producing shells with little to no vacuoles, in the future we hope to use this mass transfer model to better understand the processes which govern vacuole formation.

4.4.1. PRODUCT SHELL SPECIFICATIONS AND QUALITY

To make large target quality shells, we first had to define a goal with which our success could be measured. Working with Rich Stevens of General Atomics and Mike Mitchell of Los Alamos, we agreed on specifications for diameter, sphericity, non-concentricity, wall thickness, vacuole content, and surface finish (Table 4-3).

Table 4-3
Shell Specifications and Measurement Methods

Property	Method	Specification	Precision
Diameter	Locate edge of image	$1.00 \text{ mm} < \text{Diameter} < 1.25 \text{ mm}$	$\pm 0.01 \text{ mm}$
Wall Thickness	White light fringe shift	$5 \text{ }\mu\text{m} < \text{Wall Thick.} < 15 \text{ }\mu\text{m}$	$\pm 0.5 \text{ }\mu\text{m}$
Non-Concentricity	White light fringe center offset	$(W_{\text{max}} - W_{\text{min}})/W_{\text{avg}} < 5\%$	$\pm 2\%$
Sphericity	Locate edge of image	Minor/Major Axis $> 98\%$	$\pm 1/4\%$
Vacuoles	Scan focus through shell	0 Count $> 5 \text{ }\mu\text{m}$ $2 \text{ }\mu\text{m} > 20$ Count $> 5 \text{ }\mu\text{m}$	Single View
Surface Finish	Scan focus through shell	0 Count $> 1 \text{ }\mu\text{m}$	Single View

After attacking the problem as described below, we were able to achieve almost all of the specifications outlined above. Table 4-4 lists the actual values, including the best, we were able to achieve during FY94. The only two specifications we did not meet were diameter, where we actually made our shells slightly too large, and surface finish, where we have not been able to eliminate all debris.

Table 4-4
The Average and Best Values Obtained During FY94 Using the
Controlled-Mass Microencapsulation System

Property	Average	Best
Diameter	1.4 mm	1.38 mm
Wall Thickness	11 mm	—
Non-Concentricity	<5 %	0 %
Sphericity	>98.5%	>99 %
Vacuoles ($\geq 5 \mu\text{m}$)	0	0
(2-5 μm)	~10 – 20	0
Debris	10 's	Several

4.4.2. CONTROLLED-MASS MICROENCAPSULATION

At the end of the FY93, we had demonstrated the controlled-mass nature of the triple-orifice droplet generator. During FY94, we made modifications to both the droplet generator and the controlled-mass system which further improved control of the size and wall thickness of product shells. The single biggest change was the replacement of the stainless steel third orifice with a glass capillary approximately 18 in. long. The reason for the extreme length of the capillary was to deliver the newly formed “wet” shells to a rotating round bottom flask instead of the conical column previously used for solidification. Figure 4-7 shows the new configuration used for processing shells.

The operation of this new assembly is very similar to the previous one, with one major difference; the diameter of the shells is no longer controlled by the flow rate in the third, or stripping, orifice. Instead, the diameter of the product shells is controlled by the inside diameter of glass capillary. The shells form and increase in size until the diameter

of the shell is slightly larger than the inside diameter of the capillary. At this point, the drag on the shell increases dramatically and it is stripped from the droplet generator. During operation, we typically produce 3 shell preforms per second. As before, the wall thickness is determined by the relative flow rates of the interior water and the polymer solution orifices. Figure 4-8 characterizes the diameter of batches of shells produced with two different capillaries. The diameter distribution is extremely tight, varying only 0.01 mm. The wall thickness variation ranges as much as 10% to 20%, which corresponds to differences of only a few microns.

4.4.3. SPHERICITY AND CONCENTRICITY

Shell sphericity and concentricity were particularly difficult. In earlier processing experiments using the conical column, we were not able to meet the specifications for these properties. After changing to the rotating round bottom flask (suggested by Diana Schroen-Carey of WJSA) and modifying the composition of the starting solution, we successfully found operating conditions which allow us to produce shells which are both spherical and concentric.

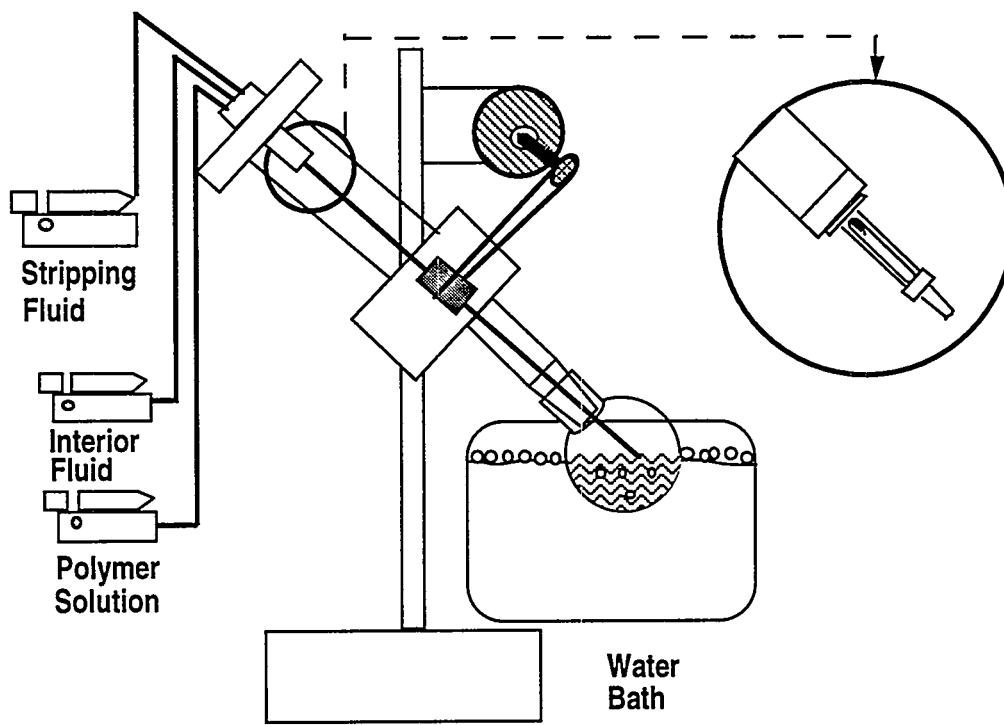


Fig. 4-7. Configuration of the controlled mass microencapsulation equipment.

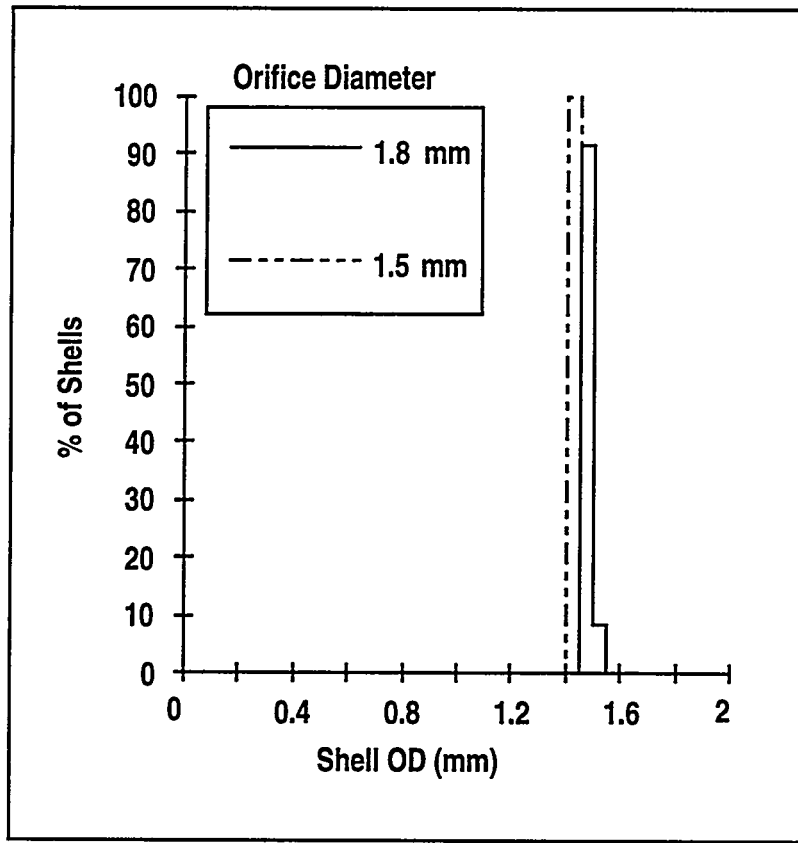


Fig. 4-8. Diameter and wall thickness distribution for two different sizes of glass third-orifice capillaries. Diameters are within 1% in size, while wall thicknesses are within 10%.

We believe that density mismatch between the inner water and the polymer solution wall is the major cause of non-concentricity in microencapsulated shells. To ascertain the most important time during the solidification process, we performed a variety of experiments and utilized the mass transfer model. The first experiment was to measure the polymer concentration the wall of the shell must reach before any non-concentricity is "frozen" in place. We determined this by measuring the viscosity of polymer solutions at a variety of concentrations and temperatures. At 40% polymer in our typical solvents, we found that bubbles did not rise over a period of several hours, indicating that the mobility of the interior fluid would be limited at this concentration and viscosity. The next experiment was to monitor a single shell as it solidified and to watch the non-concentricity as a function of time. We did this by making a long, thin column into which we could inject single shells and videotape them with a hand held camera, and analyzing the footage using frame grabbing software. We were able to measure the inside and outside diameter of each shell as a function of time. In addition, we were able to observe

the relative offset of the interior water sphere with the polymer solution wall. Using our mass transfer model, we predicted the rate at which the wall would shrink. Figure 4-9 shows the experimental measurements and the prediction from the mass transfer model using no adjustable parameters. The data and the prediction correspond almost exactly. Because of the chemical composition of the polymer solution we use to process shells, at some point in time the interior water and the polymer solution are density matched. During our video camera experiments, we observed this point in the solidification process at approximately 300 seconds. Figure 4-10 shows frames from the video tape in which the densities were matched. Notice that the interior water drop oscillated between the top and the bottom during this time, indicating nearly perfect density matching. These experiments gave us insight into the processes which contribute to concentricity.

After implementing the necessary changes, we were able to produce shells with sphericities and concentricities which meet the specifications outlined previously. Figure 4-11 shows interference photomicrographs of 1.5 mm shells produced using the droplet generator.

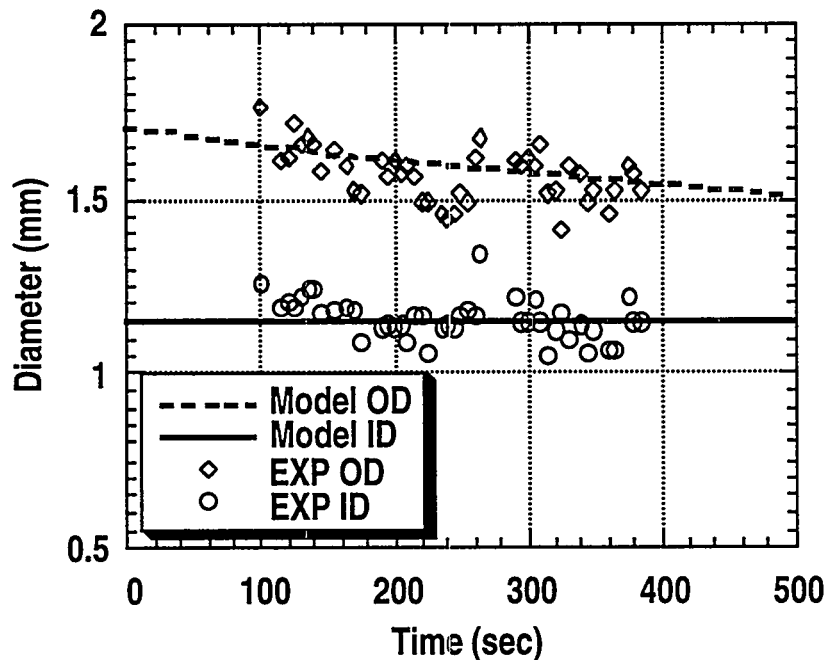


Fig. 4-9. Inside and outside diameter versus time for a shell monitored with videotape during the solidification process. The curves show the prediction from the mass transfer model.

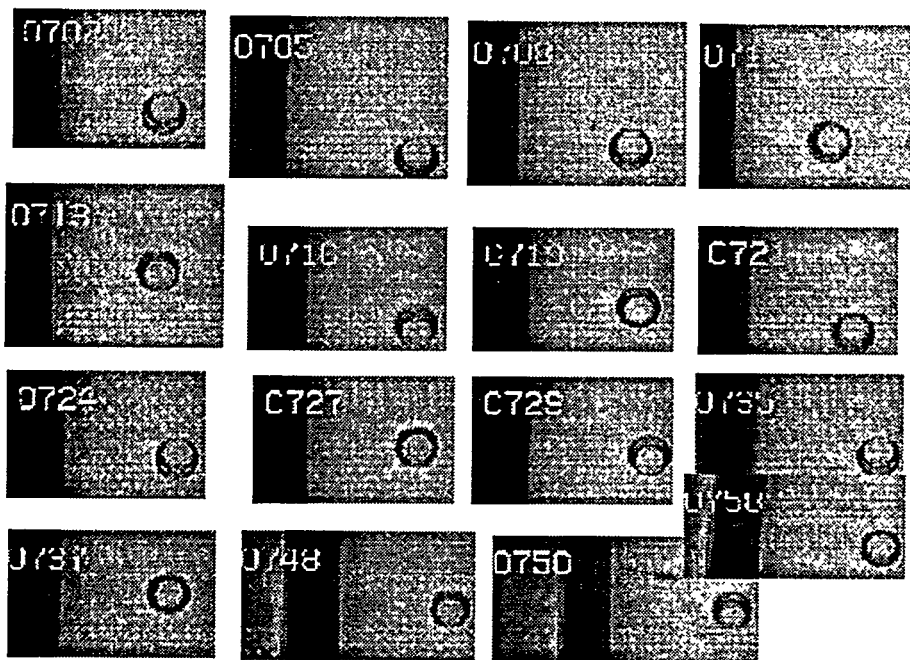


Fig. 4-10. Frames of video footage taken near 300 seconds into the solidification process. The interior water drop can be seen oscillating between the top and the bottom as it nearly matches the density of the polymer solution.

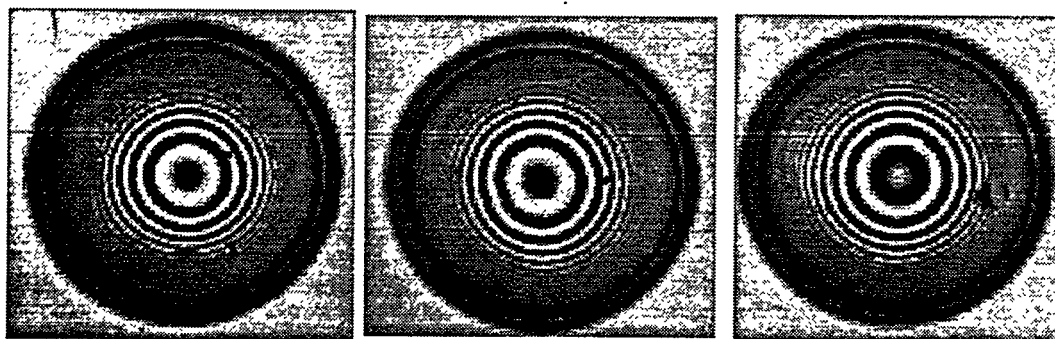


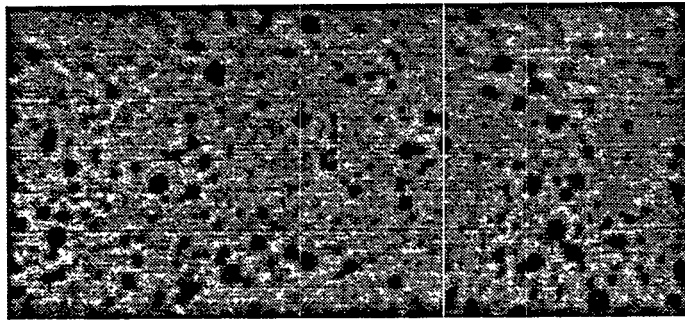
Fig. 4-11. Interference photomicrographs of 1.5 mm diameter shells produced using the controlled mass microencapsulation.

4.4.4. VACUOLE REDUCTION

Vacuole reduction proved to be one of the most daunting of tasks. Unfortunately, it is nearly impossible to watch vacuoles form. At the Mid-Year review, we had made enough progress on sphericity and concentricity to begin working on vacuole reduction. After visiting the University of Rochester and working with Steve Noyes, we began to experiment with toluene and dichloroethane as solvents. When we first began using these

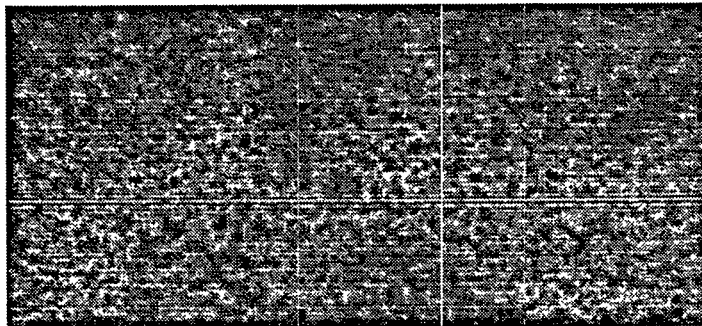
solvents, we did not observe a significant decrease in the vacuole content. However, after switching to anhydrous solvents to prepare our polymer solutions and filtering these solutions two times through 0.2 micron filters we saw significant improvement. The number of vacuoles observed which were greater than 5 microns in size dropped to nearly zero. We attribute this to elimination of nucleation sites for phase separation. Late in the fiscal year, by minimizing the wall thickness and the number of shells in each run, we succeeded in producing shells that met the vacuole specifications. Figure 4-12 shows the progression of vacuole reduction during the second half of the fiscal year.

>5 μm Vacuoles



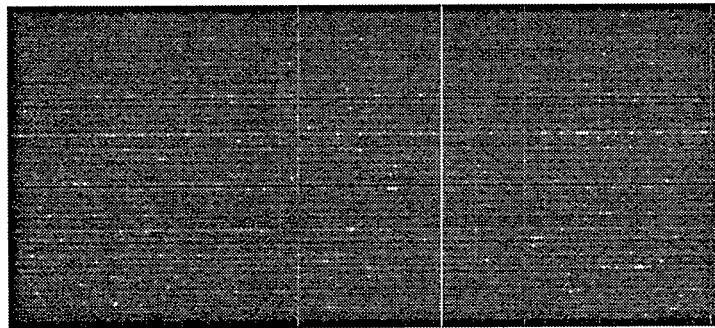
Apr 1994

~2 μm Vacuoles



Aug 1994

No Vacuoles



Sept 1994

Fig. 4-12. Photomicrographs (1000x) of 1.5 mm diameter shells showing the reduction in vacuole content during the fiscal year. Only a small portion of the shell is shown due to the magnification.

At this point in time, we do not fully understand the process of vacuole formation. Empirically, we have been able to eliminate most vacuoles. To reduce this to practice, a better understanding of vacuole formation would be very advantageous.

4.4.5. MASS TRANSFER MODELING

To better understand the physical processes governing solidification and increase chance for success, we modeled the transport processes which occur during the formation and hardening of microencapsulated shells. The goals were two-fold — to model the composition of the shell wall as a function of time which can be related to density and viscosity; and to use the results of this model, in conjunction with phase equilibria data, to predict the formation of vacuoles. During the fiscal year, we completed the mass transfer model, but not enough phase equilibria information was available to successfully predict the formation of vacuoles.

The most important unknowns in the model are the mass transfer coefficients, which determine the rate at which each of the solvents escapes to the surrounding water, and the diffusivities of the solvents in the polymer solution, which determine the rate at which the solvents can reach the outer surface of the shell. We measured the mass transfer coefficients of each of the solvents by encapsulating shells in a known volume of fluid, extracting small aliquots of the water, and subsequently measuring the solvent composition using gas chromatography. Figure 4-13 shows the data used to measure the mass transfer coefficients of each of the solvents used during the year, except for toluene, where we used the value for benzene in the model. For the diffusivities of the solvents within the shell wall, we used estimates from literature correlations.

To confirm the validity of the model, we performed several test cases. The first comparison was to compare our prediction of the diameter as a function of time with that measured as described in Section 4.4.3. In a related comparison, we compared our prediction of the time of density matching with that measured experimentally (see Fig. 4-10). Figure 4-14 shows the predicted density of the shell wall and the core as a function of time. The predicted time of density matching exactly corresponds to that measured in the video footage, indicating the predictive accuracy of the model. It is important to realize that these curves are not fits to data, rather they are predictions with no adjustable parameters other than the diffusivities and the mass transfer coefficients which we measured.

$$\text{Wt}\% = -\exp\left(-\frac{A k_m}{V} t + \ln \text{Wt}\%_{\infty}\right) + \text{Wt}\%_{\infty}$$

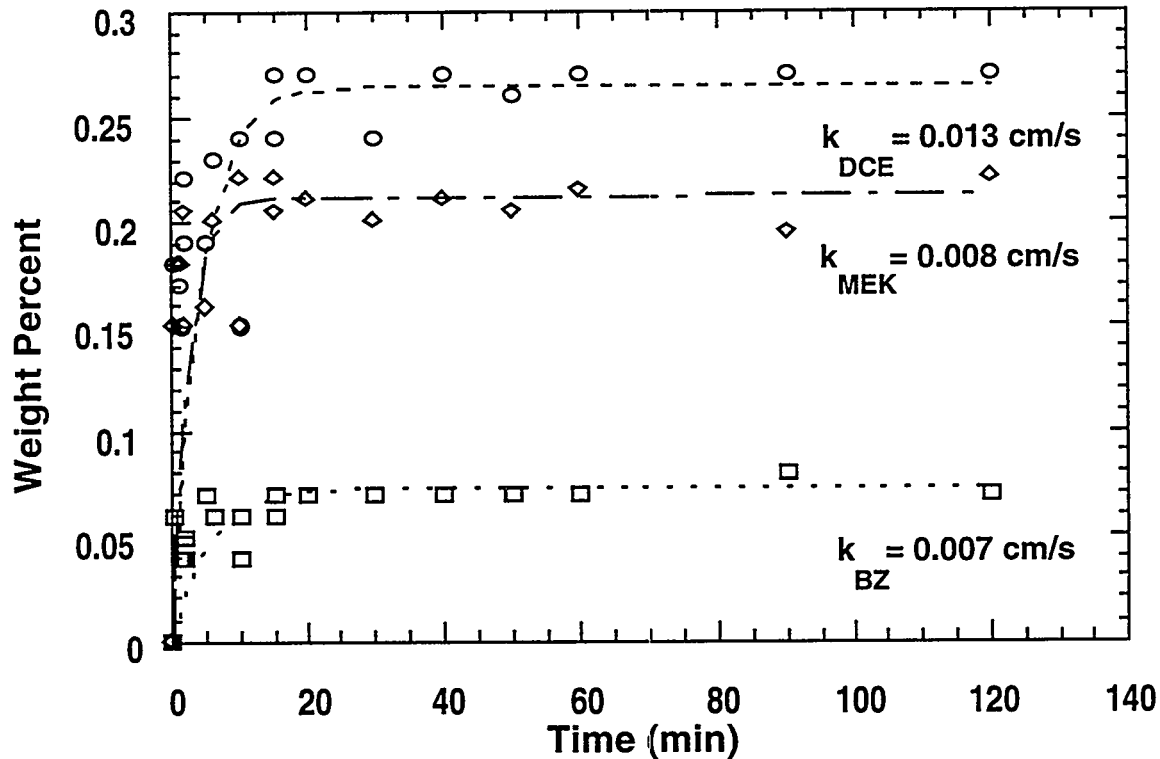


Fig. 4-13. Data and curve fit used to measure the mass transfer coefficients used in the model.

In addition to predicting the density of the shell for concentricity optimization, mass transfer modeling can be used in conjunction with phase equilibria information to predict the formation of vacuoles and to make 'test runs' of processing schemes to avoid the undesirable two phase region. During the year, not enough phase information was available to successfully use the model for vacuole abatement. We did, however, incorporate the ingress of water into the shell wall. This is important because water is a non-solvent and is the likely cause of phase separation through its modification of the solvent power of the mixture. Figure 4-15 shows the predicted concentration of water in the shell wall as a function of time. This information can be cross-plotted on a phase diagram, showing the time and composition at which the shell wall enters the two phase region where vacuole formation occurs. Details such as the viscosity and the depth of entry into the two phase envelope will affect the quantity, size, and distribution of vacuoles.

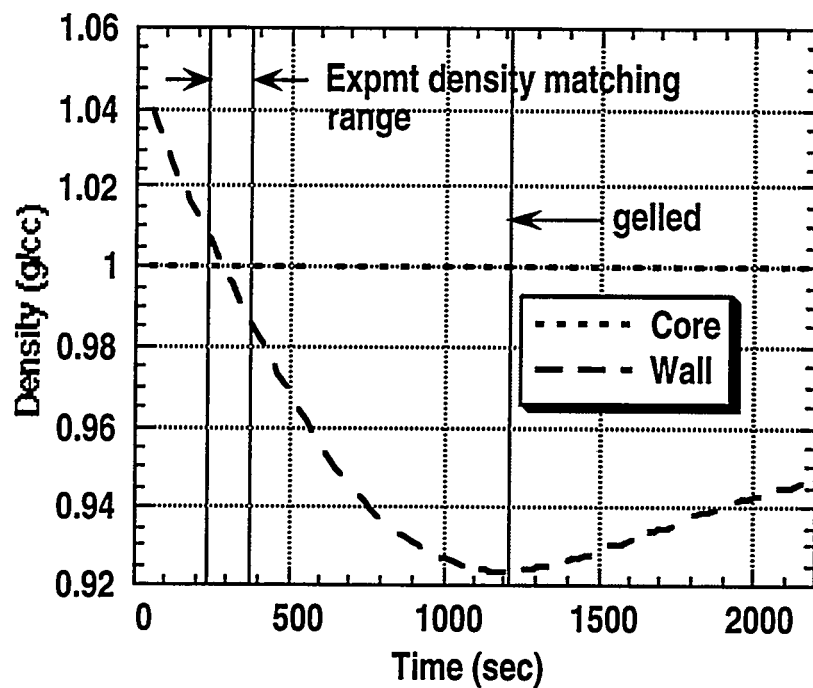


Fig. 4-14. Predicted density of both the wall and the interior fluid as a function of time. When the curves cross, density matching is achieved, minimizing the driving force for non-concentricity.

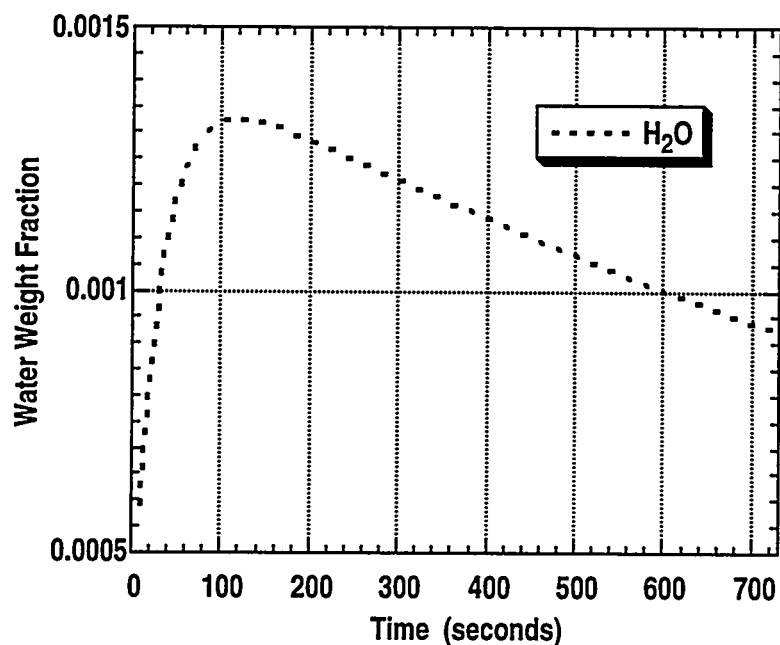


Fig. 4-15. Predicted water concentration in the mandrel wall as a function of time.

For further information, please contact Dr. D. Nelson (STI).

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SECTION 5

CRYOGENIC SCIENCE AND TECHNOLOGY DEVELOPMENT

5. CRYOGENIC SCIENCE AND TECHNOLOGY DEVELOPMENT

The goal of the cryogenic layering program is to conceive and demonstrate viable ways to generate and characterize uniform cryogenic layers.

5.1. BETA LAYERING EXPERIMENTS

Beta layering experiments with GA support at LANL have concentrated on the production of smooth solid D-T surfaces ($1.7\text{ }\mu\text{m rms}$) on layers $100\text{ }\mu\text{m}$ thick, and the development of techniques to make qualitative and quantitative improvements in surface roughness data. Two sets of experiments were performed to precisely measure solid surface finish, then improve the surface quality by a process akin to annealing. Additionally, two new cells were designed to help produce a less distorted view of the D-T solid layer than is possible with our current cell, and yield normal equilibration rates (27 min. for a 50-50 D-T mixture) instead of the 11 h measured for current experiments.

The production and measurement of high quality D-T solid surfaces requires that the solid be as free from defects as possible, and that the optical path provide as undistorted a view of the layer surface as possible. At temperatures near the triple point, defects in the D-T solid are dominated by the formation of ^3He bubbles (from beta decay in tritium) after about 4 hours of equilibration time [Ref. 5-1]. Experiments at LANL have shown that it is possible to improve layer quality by equilibrating at temperatures near the triple point (19.79 K), but that it is necessary to equilibrate the solid layer at a rate much faster than the 11 hours measured for these experiments. Short equilibration times minimize ^3He buildup in the solid layer which can roughen the surface and produce a layer on the cell front window that is not optically clear. In addition, precise measurement of the solid surface finish requires distortion free images of the D-T surface. Optical distortions are produced by both the elements in the optical train as well as solid D-T on the cell window.

The experiments described here will show the results of our attempts to anneal the layer to obtain a smoother surface finish, as well as the effects that cell morphology and

contaminated fuel can have on the solid surface finish — or more precisely, the effects these features can have on our ability to accurately measure the actual surface roughness.

5.1.1. ANNEALING EXPERIMENTS

Normally, the solid layer equilibration proceeds with a rate constant of 27 minutes for a fresh 50-50 DT mixture. Layer equilibration is complete in about two hours. However, the D-T cell and fuel configurations for these experiments have unusual features that contribute to increasing the rate constant to over 11 hours. First, the re-entrant cell geometry effectively forces the beta-layering process to proceed through an orifice which may slow the beta-layering process and lead to longer equilibration times. The other unusual feature apparent for these experiments involves our D-T fuel composition. Ordinarily, a D-T mixture contains D_2 , T_2 , and D-T molecules. However, an analysis of our D-T shows that 4% hydrogen, in the form of H-D and H-T molecules, is also present in the mixture. The presence of this hydrogen (triple point temperature = 14 K) can slow the beta-layering process by introducing a non-condensable gas in the vapor space which impedes the mass transport of D-T. The hydrogen can also lower the melting temperature of the mixture, making it difficult to make precise measurements of layer surface roughness in the temperature regime of interest.

Original Cell Geometry. The first series of experiments were performed to investigate the technique of layer smoothing by annealing, and results showed that it is possible to smooth solid layers by equilibrating near the triple point. These experiments also showed that apparent surface roughness increases as a result of long equilibration times and build-up of solid D-T on the cell front window. These long equilibrations produce ^3He bubble defects that can produce both surface roughness and distortions in the optical train.

We ran the first half of the experiment for over 300 hours, making four temperature changes along the way. Our first temperature increment, from 18.48 K to 18.7 K, was made at just over 70 hours and produced an immediate improvement in the layer surface finish over several hours, as shown in the surface roughness evolution graph in Fig. 5-1. After about 8 hours, the layer appeared to degrade, recover, then degrade again over a 60 hour period. Initially, we thought that this apparent layer degradation was again due to layer softening and we began a process of lowering the temperature in three degree increments. In all cases, the layer surface finish improved initially, followed by an

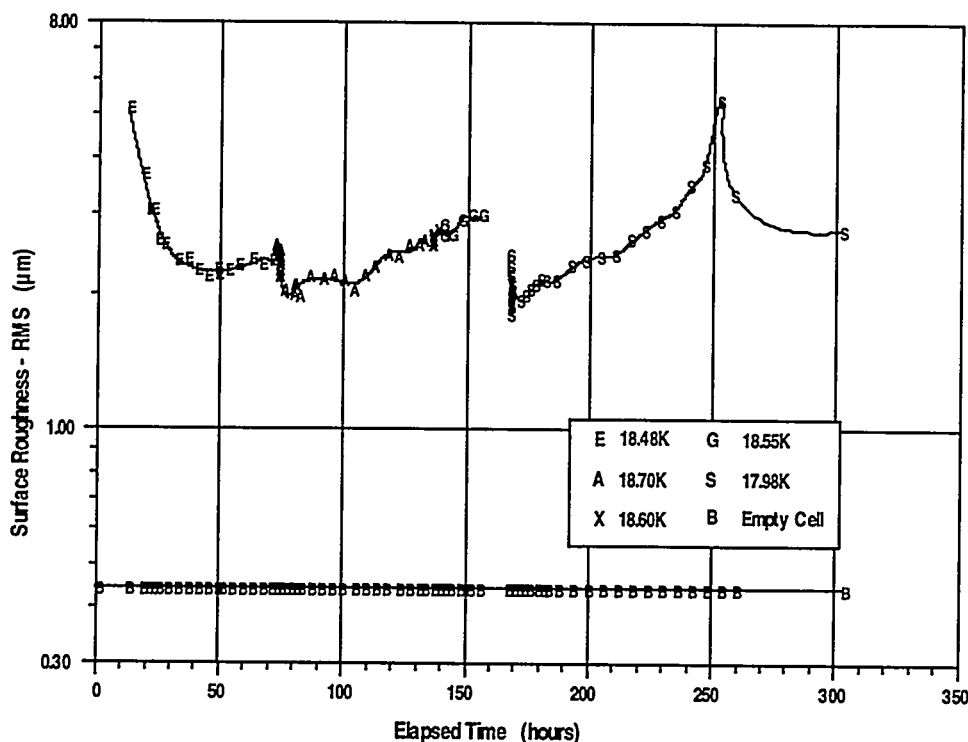


Fig. 5-1. Surface roughness evolution for the temperature stepped 105 μm D-T solid layer. The large roughness spike seen in the 17.98 K layer is caused by a ^3He bubble crossing the field of view.

apparent slow degradation (It is interesting that the layer always improved at first, for temperature changes in either direction. It seems that each temperature change forces a temporary reconfiguration of the layer surface, after which a more dominant process takes over).

We began our observations of the cell front window during the apparent degradation of the 18.7 K layer and noticed that dark spots and circles were forming in the solid D-T layer on the window inner surface which slowly migrated across the window. These spots, or bubbles, would occasionally cross the field of view for the solid D-T layer on the cell inner bore. Such bubbles form as the result of tritium beta decay to ^3He and we suspected that, when formed in the window D-T layer, they might blur the image of the inner bore D-T layer. While observing the layer equilibration following the temperature change from 18.55 K to 17.98 K (a lowering of T), we allowed the layer to evolve for over 134 hours. We assumed that the layer would continue to degrade throughout its evolution and were surprised to observe this large spike in the surface roughness profile. Because we had been observing the bubbles form and migrate across

roughness profile. Because we had been observing the bubbles form and migrate across the cell front window, we deduced that this spike was likely one of those bubbles passing through the field of view of the layer edge.

In order to test this hypothesis, we examined the front window and layer images during the time of the roughness spike, side by side, to see if a bubble in the window layer was moving through and obscuring the bore solid layer. Additionally, we plotted the layer surface contour changes during the spike to see if there were any significant changes in the contour at certain radial positions in the cell. Figure 5-2 shows the results of this investigation, and along with Fig. 5-1, is clear evidence that bubbles passing through the field of view can distort the image of the layer edge and increase the apparent surface roughness of the solid layer.

Figure 5-2 clearly shows the impact of front window bubbles on the measurement of layer surface roughness - note the distortions in the layer in regions where large bubbles occur on the front window. The images of Fig. 5-2(a) show that the layer edge is smeared at the precise position of a massive bubble on the front window at about the 1:30 position, which can be seen in the images of Fig. 5-2(c). The polar plots of Fig. 5-2(b) show the perturbation passing through the layer at precisely the same position shown in Fig. 5-2(a), and time as shown in Fig. 5-1. We conclude that the roughness spike seen in Fig. 5-1 is indeed a ^3He bubble, or bubble complex, moving through the field of view for the layer edge.

The roughness quality of the solid layer on the inner 2 mm bore may also be affected by ^3He bubble formation, but this cannot be determined without a clear view of the layer surface edge itself. The effects of ^3He buildup and the migration process are particularly acute for this experiment because of cell morphology and experimental conditions. Cell geometry as well as the presence of hydrogen in our D-T mixture both contribute to slowing the rate constant considerably, which allows time for the ^3He bubbles to accumulate and migrate. This process occurring in the solid layer frozen on the front window inner surface makes it impossible to get a clear look at the D-T layer surface on the inner bore of the cell. We will address these problems associated with cell geometry and front window clarity for our next experiments, using upgraded D-T cells with improved morphologies and front window characteristics.

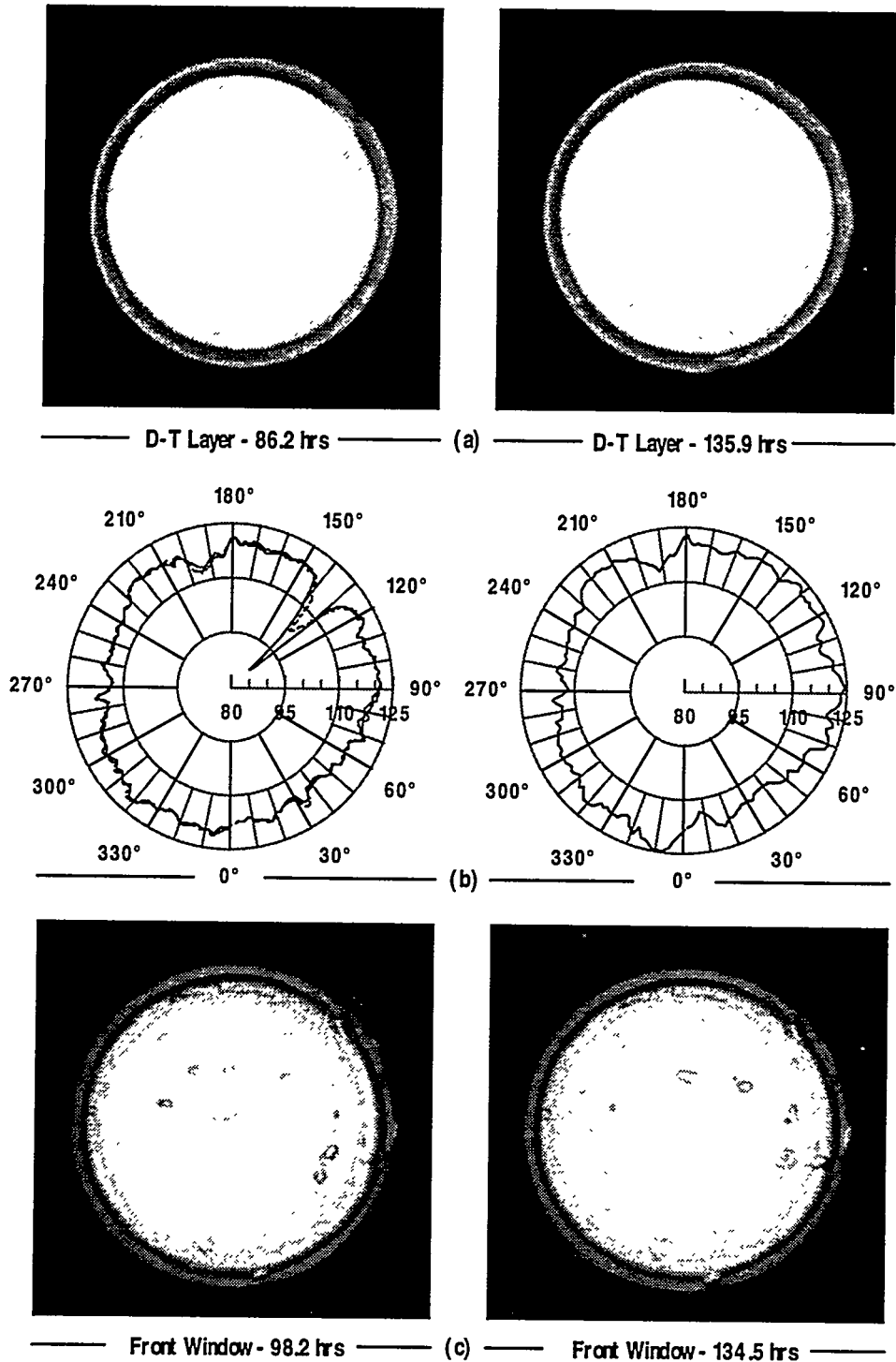


Fig. 5-2. Images of the 105 μm D-T solid layer at 17.98 K. (a), and front window images within the same time frame (c). The polar plots in (b) show the layer surface morphology at the times shown for the images above them (a), with layer thickness plotted over cell angular position. The dotted line in the polar plot at the left is the layer morphology at the time of the front window image shown directly below (c).

Modified Cell Geometry. The next experiment was performed using a glass hemisphere, designed by Mapoles at LLNL, and was intended to produce an undistorted view of the D-T solid layer surface formation in spherical geometry. The hemi was sealed by a thin, flat glass window of low thermal conductivity, with attached resistive heaters that prevent the formation of D-T solid on the window, providing an unperturbed view of the solid layer inside. Unfortunately the hemi developed a sizable leak, the magnitude of which became apparent during testing with liquid D₂, and so the experiment was abandoned. However, the images taken during this D₂ liquid fill provided interesting data from which we were able to measure the liquid surface finish.

Figure 5-3 shows the results of this liquid surface finish measurement and compares it to the surface roughness that was measured for an earlier 125 μm solid layer. These graphs show that with the exception of an expected P1 (off center) defect, the liquid surface is smoother than the solid by nearly an order of magnitude (0.33 μm vs. 2.2 μm).

5.1.2. CONCLUSION

These experiments clearly demonstrate that it is possible to smooth D-T solid layers by the annealing process. We were not able to determine just how smooth the layers can get because of the limitations imposed by long equilibration times and consequent ³He bubble buildup in the solid layer — both in the cell bore and on the front window. This ³He buildup on the cell front window creates distortion in the optical path and a noise floor that cannot be penetrated, thereby limiting the accuracy of the surface finish measurements. The next generation of D-T cells will have heated front windows and be using uncontaminated D-T, thus providing a much shorter equilibration time and a solid free, lower distortion optical path. Additionally, we are working on a technique to remove distortions caused by light propagating through the optical train (point spread function), called maximum entropy deconvolution. This should allow us to determine if solid D-T layers will be able to meet NIF roughness specs of 0.5 μm rms over a 6 mm length scale.

For further information, please contact J. Simpson (GA at LLNL).

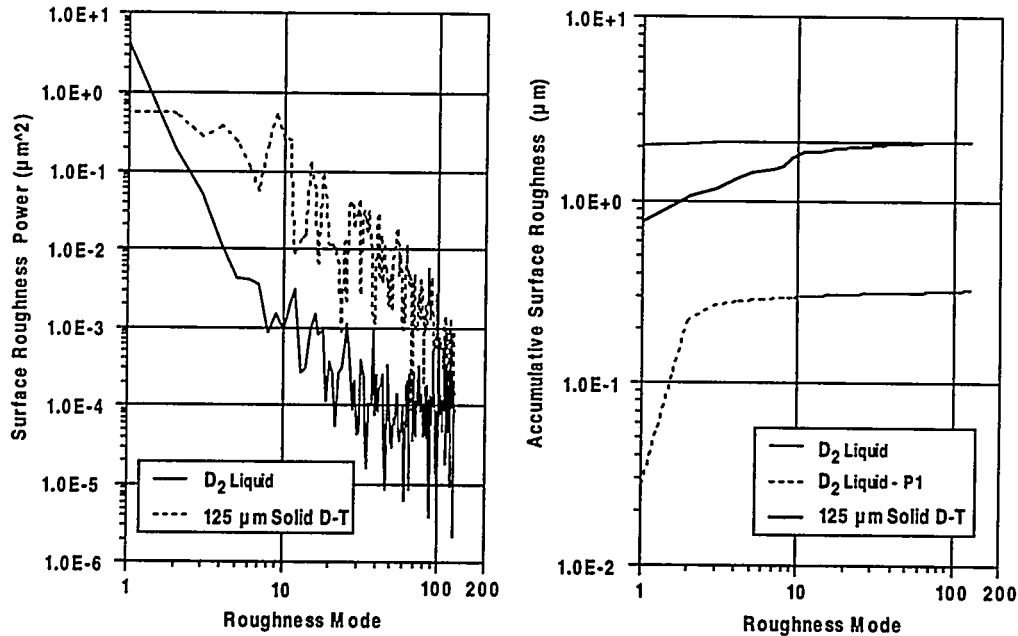


Fig. 5-3. FFT plot of D₂ liquid layer inside 2 mm diameter glass hemisphere (lower plot), and 125 μm D-T solid layer inside 2 mm diameter copper cylinder showing significantly smoother liquid layer at mode >P1. (b) Accumulating sum of individual Lagrangian modes terminating in the final ms surface roughness for the D₂ liquid layer (lower plot) and 125 μm D-T solid layer. Liquid is nearly an order of magnitude smoother than the solid layer.

5.2. CRYOGENIC LAYERING TECHNOLOGY

Cryogenic layering technology was centered on two tasks. One was the investigation of deuterium flow rates through micro-tubes which are to be used to fill ICF targets. The other was developing 1-D NMR imaging NMR probe that will be used to examine beta-layered D-T samples for isotopic fractionation. Roy Mangano of General Atomics supported the micro-tubes experiment. Neil Alexander supported the NMR experiment.

5.2.1. MICROTUBES

The aim of this experiment is to determine if microtubes would be suitable for use as fill tubes for ICF targets. Theoretical flow rates for deuterium through micron and sub-micron inside diameter tube are enormously sensitive to assumptions made about the boundary conditions of the flow. Both acceptably short and unacceptably long fill times can be obtained by adjusting the boundary conditions, thus experimental data is required to resolve the utility of micro fill-tubes.

An apparatus for measuring flow rates through micro-tubes at low temperatures was designed and the components procured. A CRADA was established with Edward Air Force Base — Phillips Labs for procurement on the microtubes. A set of mounted micro-tubes was ordered with an i.d. range of $0.5\ \mu\text{m}$ to $10\ \mu\text{m}$. The tubes were not delivered in FY94 as planned, and the effort has been put on hold. When these microtubes are received, this experiment will resume.

The design of the apparatus is illustrated in Fig. 5-4. The microtubes are mounted on a fixture that is placed inside a chamber. The chamber is cooled in a liquid helium dewar. A temperature controller maintains the temperature of the fixture at the desired level. Plumbing to the chamber allows the strength of the thermal exchange gas link to the liquid helium to be adjusted. Keller PSI cryogenic pressure transducers are placed immediately before and after the micro tube. This allows measurement of the differential

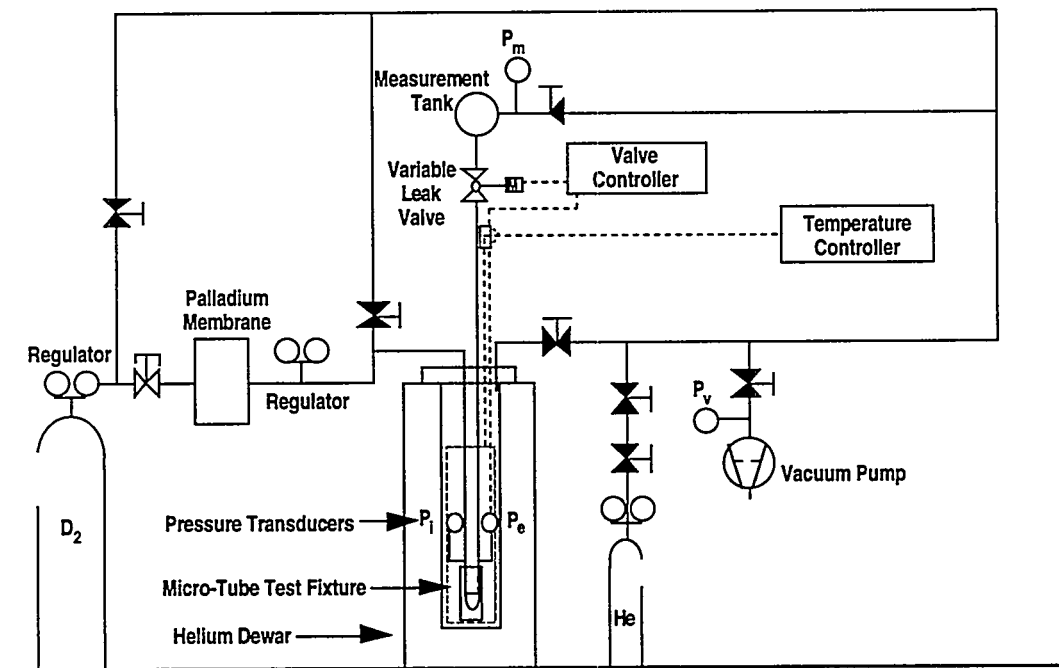


Fig. 5-4. Microtube flow rate apparatus.

pressure driving the flow. Bottled deuterium gas is purified by running it through a heated palladium membrane before it enters the cryogenic chamber. Additional filtering is accomplished with a cryogenic $0.5\ \mu\text{m}$ frit filter placed in the line ahead of the microtube. A pressure controller connected to a variable leak valve down stream from the micro-tube, and a large control volume, are used to adjust the down stream pressure to prevent

two-phase flow from occurring in the microtube. The large control volume can also be used to measure the total mass of gas passed through the microtube.

For further information, please contact Roy Mangano (GA).

5.2.2. SPATIALLY RESOLVED NUCLEAR MAGNETIC RESONANCE

Inertial confinement fusion targets require a fuel layer that is both uniformly thick and uniformly dense. If a layering process should improperly fractionate the targets' D-T fuel, the resulting fuel layers would have non-uniform density. A spatially resolved (1-D), doubly tuned NMR spectrometer will be used to investigate fractionation of D-T samples undergoing the beta-layering process [5-2 through 5-6].

With a doubly tuned spatially resolved NMR probe, the density of deuterons and tritons may be determined separately as a function of height throughout the sample. In-depth details of the experimental plan were given in last year's annual report [Ref. 5-7].

Last year, a feasibility study showed experimentally that solid hydrogen could be 1-D imaged [Ref. 5-7]. This year, the design of last year's probe was updated to be tritium compatible and the magnetic field gradient coil design was improved. A cartoon of the NMR probe is shown in Fig. 5-5. This new probe design was completely assembled and partially tested.

Tritium compatibility was added to the probe by providing complete secondary containment of the sample cell and fill line within the probe. A glass bulb seals the bottom of the transmission line. The sample cell and RF coils lie within the bulb. The gradient coils are wound directly on the glass bulb. The bulb is sealed to the transmission line using a copper to glass seal made by Larson Electronic Glass. The upper portions of the transmission line are sealed by potting and by glass to metal transitions. The fill line within the transmission line is glass to prevent electrical shorting. The upper portion of this glass tube is epoxied into a metal tube. The lower portion is epoxied to a fused silica capillary which will connect to the sample cell.

The magnetic field gradient coil was designed by the target field method [Ref. 5-8] to be linear to within 1% over a length twice that of the sample. To improve the speed at which the coil can be switched on and off, another gradient coil with larger diameter is used. This gradient shield coil produces a gradient in the opposite direction. The current

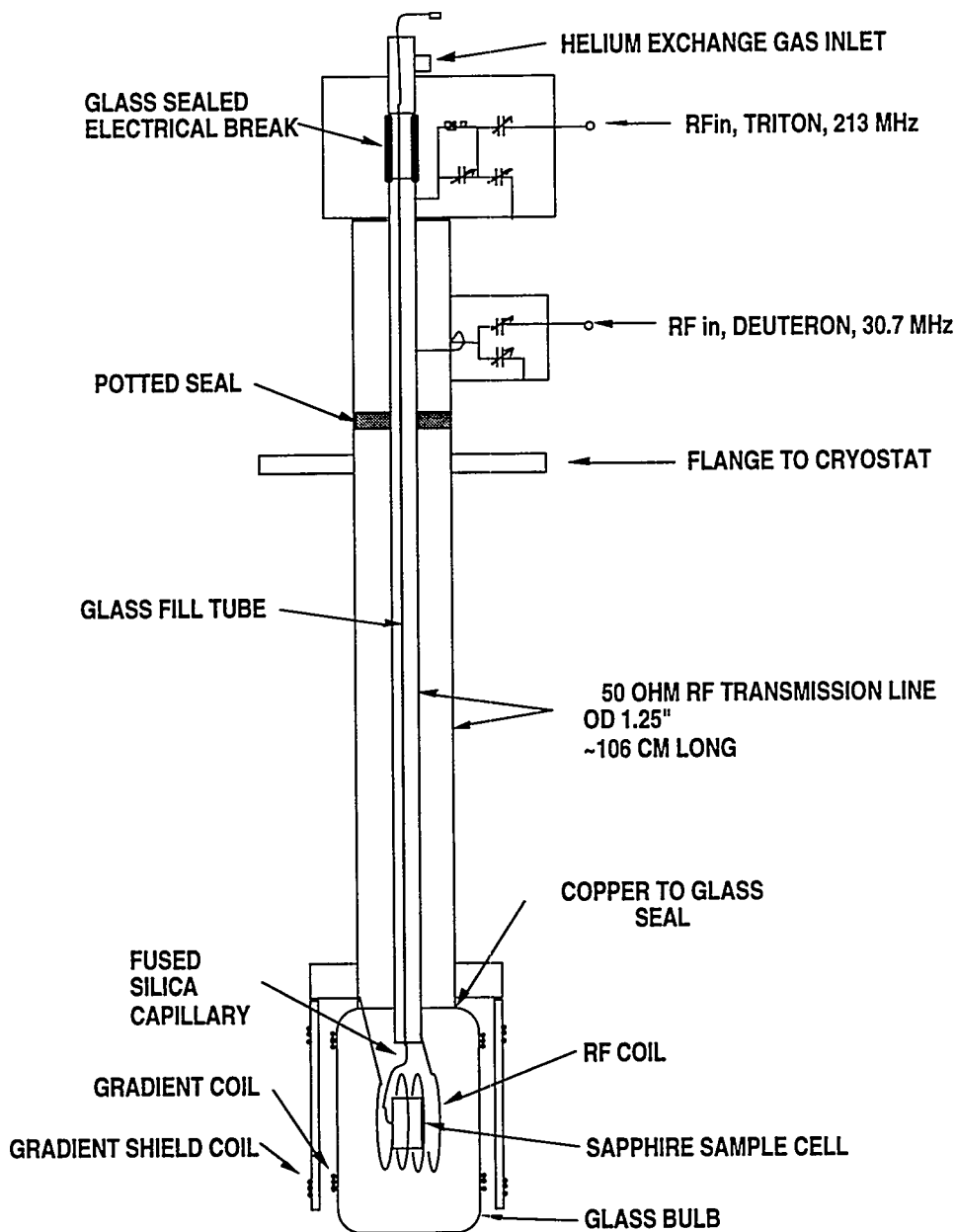


Fig. 5-5. NMR probe.

density of the shield coil is picked so that the net field produced outside both coils is zero. This reduces the gradient per ampere of current that the coils produce at the sample, but it prevents switching slow-down caused by production of eddy currents in the surrounding metal of the cryostat in which the probe resides. The shield coil has the same current density profile as the gradient coil, but reduced by a factor of the ratio of their radii squared [Ref. 5-9].

Tests of the NMR probe were performed at cryogenic temperatures in the range 3.8 K to 25 K. The probe was able to tune at both the deuteron and triton frequencies. The temperature stability while under active control was at least 0.01 K at 20 K. The helium consumption while under temperature control was one liter per hour. This performance is sufficient for the beta-layering experiment. Unfortunately, the sample cell leaked, thus no NMR measurements were performed. The leak site was located and the cell shipped back to General Atomics for repair in FY95.

For further information, please contact Dr. Neil Alexander (GA).

5.3. CRYOGENIC LAYERING DEVELOPMENT

D. Bittner, J. Burmann, and J. Sater conducted cryogenic layering experiments in collaboration with the LLNL staff. Analytical modeling support was provided by R. Bieri and M. Monsler. Plasma discharge experiments were conducted by a post doctoral fellow, C. Chen, on site at UR/LLE.

5.3.1. CLAMPED SHELL

Much work has been done on liquid layering of hydrogen in spherical containers [Refs. 5-10 through 5-13]. Most early work concentrated on layers less than 10 μm thick inside of shells typically 600 μm o.d. or smaller. Proposed target designs for UR/LLE's Omega Upgrade call for 1 mm diameter targets with 100 μm thick layers. The NIF will need even larger diameter targets. Sanchez et al. [Ref. 5-14] have done work on liquid layers inside relatively large capsules.

We undertook an experiment to determine how thick a liquid layer could be levitated inside of an Omega or NIF class spherical shell. In this experiment, a heater was placed directly on top of a shell which in turn was directly upon a copper cold finger. This design is better suited to putting a larger thermal gradient across the shell for a given average temperature than are parallel plates with heat conduction through helium gas. Disadvantages are layer non-uniformity and a fixed direction for the thermal gradient — the heater is always warmer than the cold finger. Because the heater is on top, the layering must be done for isotopic mixtures only.

Original Direct Contact Design. Figure 5-6 shows the configuration of our first clamped shell experiment. The shell is a 2 mm o.d. free standing plasma polymer layer prepared by a process described by Steve Letts [Ref. 5-15].

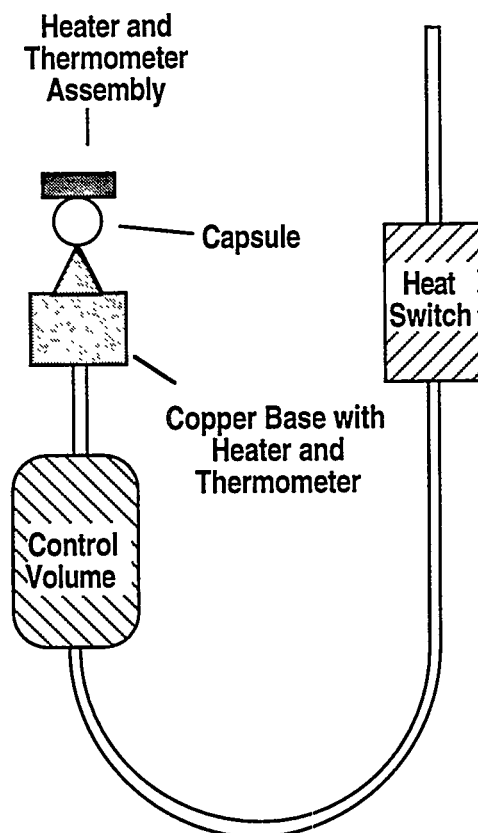


Fig. 5-6. Sketch of clamped shell experiment.

The gas mixture is introduced into the assembly downstream from the heat switch (the capsule side). When sufficient liquid has accumulated in the assembly, the heat switch is turned off. The liquid inside the heat switch then cools to about 8 K, resulting in an ice plug in the line which effectively seals the system.

The control volume is a temperature regulated cavity with approximately 100 times the volume of the capsule. By changing the temperature, we change the density of liquid in the control volume. With the heat switch closed, this density change causes excess liquid to flow into or out of our plastic capsule.

In early experiments we observed unexpected layering behavior. The fluid in the shell collected at the top, as expected. However, layering behavior was also observed at zero input power to the top heater assembly. An apparent heat leak along the wires into the heater existed. Pulsations of the liquid layer also were prominent. In fact, a stable liquid layer was observed only one time during the direct contact experiments.

Pulsation Frequency Model. An explanation was proposed for the persistent unsteady 0.3–0.5 Hz pulsation observed in clamped shell experiments. The sequence is as follows:

1. A temperature difference across the shell causes a concentration difference across the liquid-vapor interface (bubble). This gradient in surface-tension due to the temperature and concentration differences causes an upward shear-stress on the interface.
2. The upward shear-stress exceeds the retarding effects of viscosity and gravity, pulling the liquid up in a thin film on the inside of the shell.
3. The fluid accumulates in a droplet at the top, which maybe somewhat offset from top-center due to asymmetries.
4. The droplet at the top is held up against gravity by normal surface tension, until a critical size is reached.
5. When the droplet gets too heavy, it starts to fall.
6. Since the upward flow of fluid is essentially constant, the cycle repeats.

For a 2 mm diameter shell charged with H₂-D₂, a pulsation frequency of 0.5 Hz is consistent with calculations for a reasonable range of liquid film thicknesses (3–9 μm). The rise-time or time for the droplet to reach critical size (~2 s) determines the fall time (~0.015 s). The frequency of pulsation can be approximated by

$$f_a = \frac{1.55\rho g \delta^2 \tau}{\mu s \cos\phi}$$

where ρ is the liquid density, g is the acceleration of gravity, d is the liquid film thickness, t is the shear stress due to the gradient of surface tension, μ is the viscosity, s is the surface tension, and f is the offset angle of the droplet off top-center.

Three conditions have been identified for which the pulsation cannot occur. First, if the temperature difference across the shell is less than a critical value, the upward surface-tension-gradient cannot overpower gravity, the fluid will not be pumped upward, and the pulsation cannot occur. Second, if there is insufficient fluid in the sphere (small potential symmetric layers), there is less total volume of liquid H_2 - D_2 than the critical volume, and the droplet will stay at the top and not fall, and a repetitive pulsation will not occur. Third, if there is too much liquid in the shell (a small bubble), there is not enough room for a critical volume to accumulate as needed, and the pulsation will not occur.

The parameter space in which steady-state layering is desired occurs within the parameter space for which the pulsation has been observed. It remains uncertain whether steady-state layering is possible for the layer thicknesses of current interest using this technique.

Laser Heating. A new shell was mounted for this experiment. In addition, advantage was taken during the remounting of the shell to modify the probe. A laser was used in place of the previous assembly to apply heat to the top of the shell. This allowed flexibility to vary the location where heat is applied to the shell while simultaneously removing any possibility of extraneous heat leaks at zero laser power. The disadvantage of this change was the thermometer on top of the shell had to be eliminated along with the heater. This meant relying on the measurement of input laser power to determine the power flux through the plastic capsule.

The new design does not exhibit liquid oscillations when power is not applied to the top of the shell which indicates no heat leaks are present. When power is applied, the liquid oscillates just as in the previous version of the clamped shell. The total fluid content of the shell changes with laser power but can be controlled to some extent by adjusting the filling protocol for the capsule/control volume system.

Figure 5-7 shows some typical results from analysis of streak images. A streak image is composed of a series of 2-pixel wide by 1024-pixel long images taken through the center of the capsule, with the long axis parallel to gravity, and laid side by side. Streak images maximize the data collection rate and track the oscillatory behavior of the liquid layers. The x-axis in this figure is an estimate of laser energy absorbed per unit time on the top of the shell. The black squares represent the maximum thickness of a suspended oscillating layer of liquid. The hollow squares show the measured liquid layer

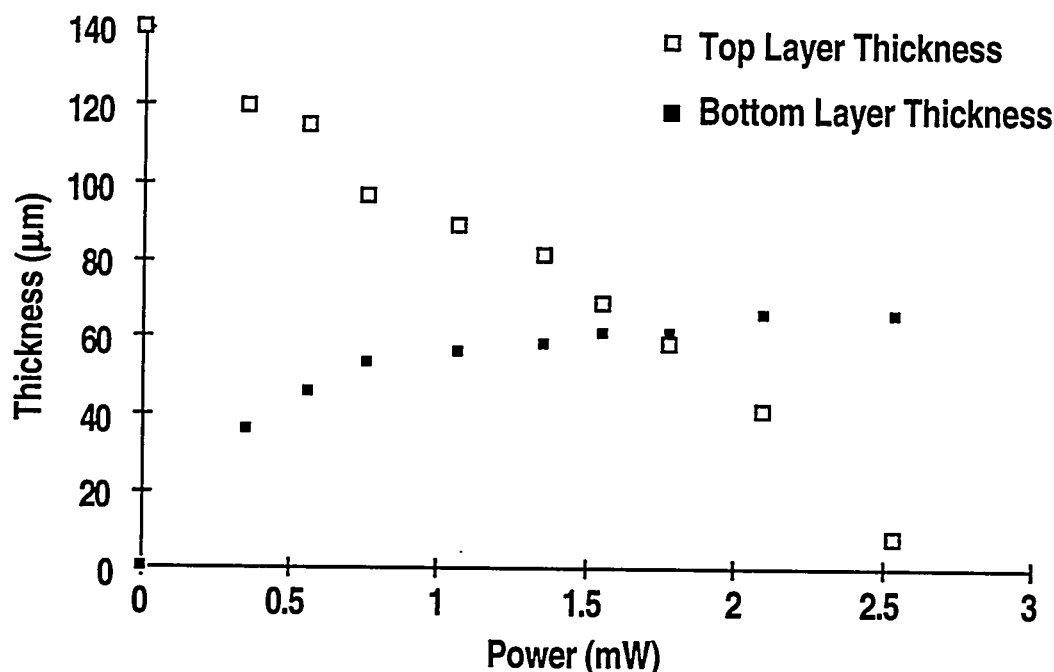


Fig. 5-7. Thickness of the liquid layers on the bottom and top of our plastic shell. The bottom temperature was maintained at 17.5 K.

thickness on the bottom of the shell. The bottom layer thickness decreases with applied power, not only because some liquid moves to the top of the shell, but also because some liquid is driven out of the shell as the power increases.

Although the analysis is not yet complete, data taken on the oscillatory behavior by recording the intensity of a few pixels at the edge of the top liquid layer readily shows an important qualitative result. A region in phase space occurs where long period oscillations of the liquid layering process is observed. In this region the period can be greater than 10 seconds, and the layers are on the order of 50 μm thick. The region of long oscillation periods was found at high incident laser power, just below a value necessary to cause the top liquid layer to disappear. Systematic mapping of the region of phase space where this behavior occurs is in progress.

Layer Shape Model. A modeling study was undertaken to determine the heating profile needed to create a uniform temperature gradient on the inner liquid surface. As a first step in this modeling process, a simplified 2-region capsule model was created. The model consists of a shell with the thermal conductivity of polystyrene and a high-conductivity center (representing the higher effective conductivity of the liquid/vapor

region). Although the model is not detailed enough to predict the required heating for a given capsule, the model has demonstrated:

1. The effects of changes in the effective conductivity of the liquid and vapor on internal capsule temperature profiles.
2. The effect of changes in the applied external temperature or heating profiles on internal temperature profiles.
3. The external heating or temperature profiles needed to create a uniform temperature gradient (and thus a uniform surface tension gradient) at the region boundary surface.

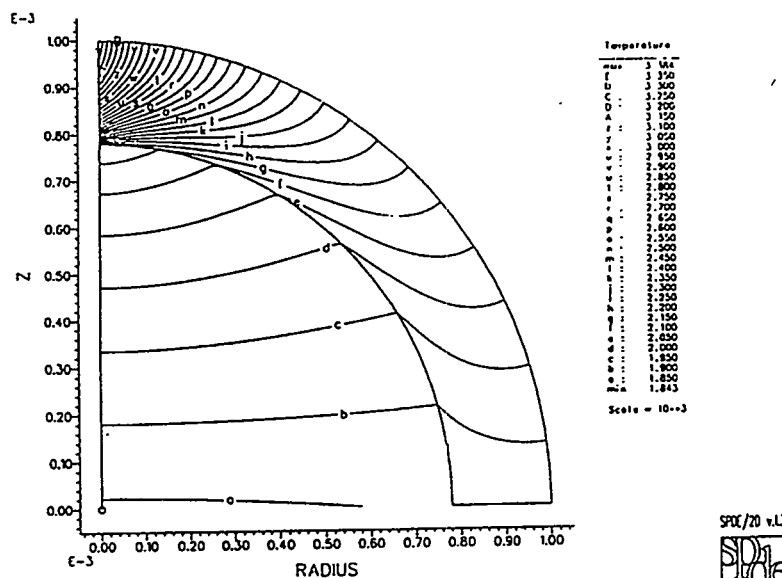


Fig. 5-8. Cos(q) heating profile. The effective conductivity of the center is 10 times that of the shell.

The temperature profile along the inner shell surface will approximate the shape of the temperature profile along the liquid/vapor interface. The relative strengths of the surface tension gradient in various parts in the capsule are seen by examining the spacing of the calculated isotherms. One would expect the experiment to have thin spots in the liquid layer, wherever this gradient is relatively low. For example, Fig. 5-8 shows a

temperature profile for a $\cos(q)$ heating profile consistent with a thick layer on top of the capsule which progressively thins upon approaching the capsule equator. Both the $\cos(q)$ and a sharply peaked exponential heating cases give profiles which have low thermal gradients as you approach the equator. For our simple model, the $\sin(2q)$, Fig. 5-9, heating profile gives the desired thermal profile.

5.3.2. UNIVERSAL CURVE

J. Sanchez of LLNL has found a correlation which seems to allow the plotting of many different thermal-layering experiments on the same graph [Ref. 5-14]. This correlation is expressed as a plot of the thermal gradient across the entire shell (in K/cm) divided by the levitated layer thickness (in μm) versus average shell temperature. The results from several different experiments, performed at the University of Illinois and LLNL, are plotted in Fig. 5-10. Generally, as the average temperature increases from 18 to 26 K, the temperature gradient required increases slightly, while the layer thickness decreases slightly. This is due to the increasing fraction of the mixture in the vapor state.

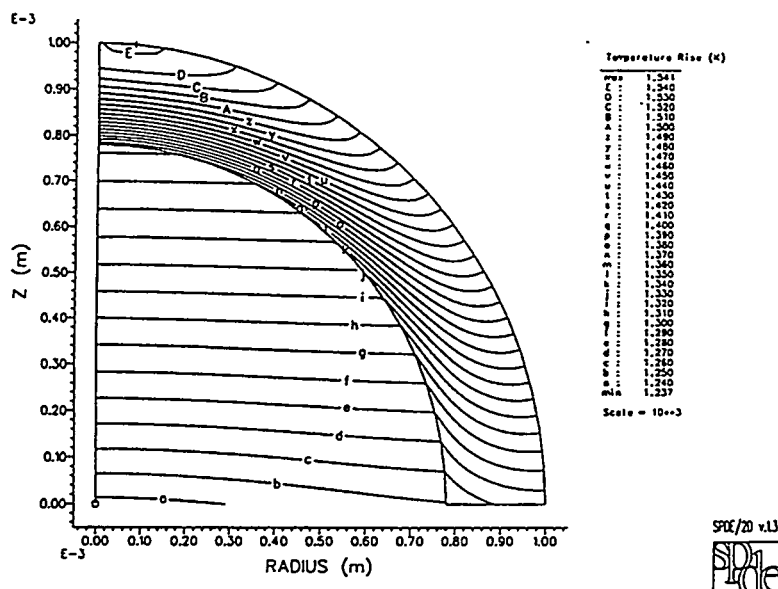


Fig. 5-9. $\sin(2q)$ heating profile. The effective conductivity of the center is 10 times that of the shell.

Some of the data veer sharply upward over a temperature range of only 1.5 K. These data points, which were taken in the "dynamic" layering experiments, seem to have an inexplicably large reduction in layer thickness as temperature increased only slightly. Perhaps, some of the fluid was driven out through the fill tube by the increasing vapor pressure. Nevertheless, data taken over a large range of shell sizes and fuel fills can be put on the same graph, in what has been termed the Sanchez Correlation. Why should a semi-universal curve like this for the thermal-layering of liquid H₂-D₂ mixtures be expected?

A simple theoretical explanation for this correlation was worked out, which results in the solid curve of the Fig. 5-10, provided that the unknown total derivative of the surface tension with temperature is the following constant value.

$$\frac{d\sigma}{dT} = \frac{\partial\sigma}{\partial T} + \frac{\partial\sigma}{\partial x} \frac{\partial x}{\partial T} = 8.0 \times 10^{-3} \frac{\text{J}}{\text{m}^2\text{K}}$$

The simple analytical model combines several algebraic equations that express the following dominant physics occurring in thermal layering:

1. A temperature difference impressed across the entire capsule causes heat to flow through the capsule.
2. The heat flow through the capsule causes a very small but essential temperature difference across the bubble.
3. This small temperature difference causes preferential vaporization and condensation of the more volatile component, the H₂, setting up a significant concentration difference across the bubble.
4. The concentration difference across the bubble causes a substantial surface-tension-gradient, embodied as an upward-directed shear-stress on the surface of the liquid.
5. This shear-stress continually lifts an inner layer of the liquid to the top of the shell, where it heats and flows downward in an outer layer adjacent to the shell.

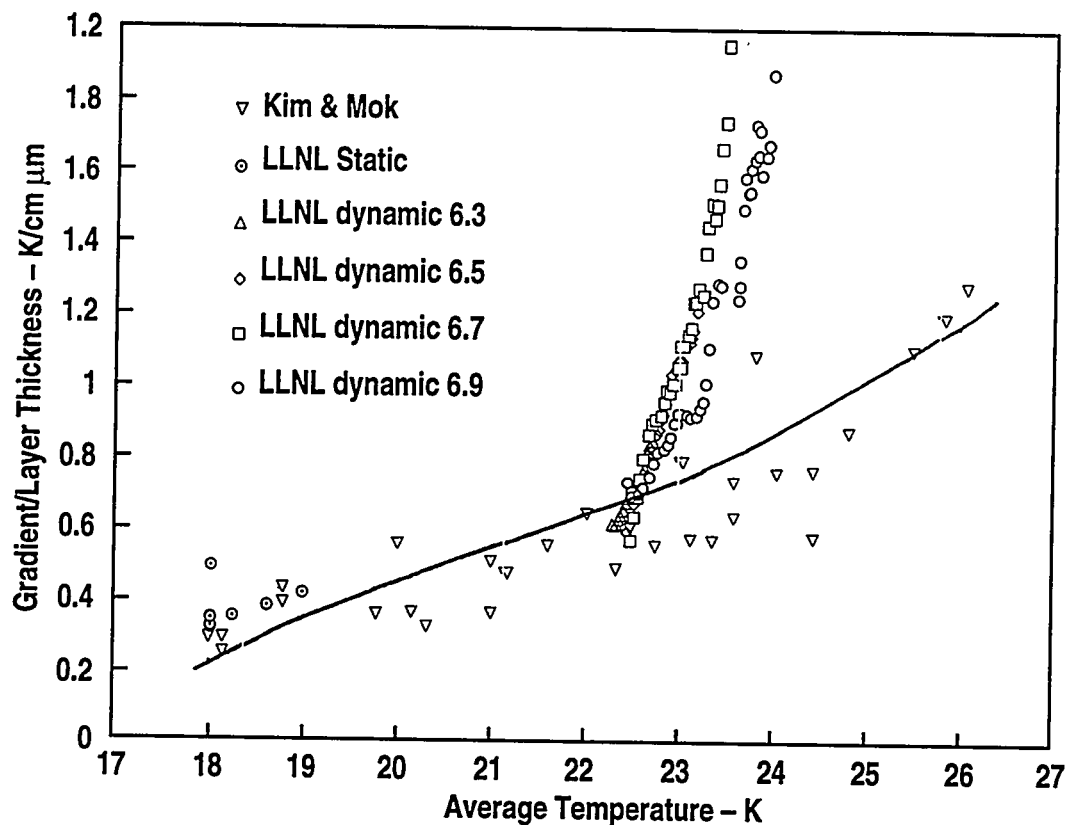


Fig. 5-10. The thermal gradient (K/cm) needed to levitate a liquid layer, divided by the layer thickness (μ m), as a function of the average temperature. (Data courtesy of J. Sanchez of LLNL, curve by M. Monsler of WJSA.)

6. The heat flow through the capsule is almost entirely carried by the flow of vapor carrying the heat of vaporization.
7. The heat flow inside the shell is nearly isothermal because liquid convection and phase change are so efficient compared to conduction.
8. The majority of the temperature change occurring across the capsule is due to the temperature drop across the shell thickness (top to bottom) as heat is conducted through the solid.

In this simple model, the heat transferred through the entire capsule is set equal to the heat carried by the mass flow of vapor times the heat of vaporization. The power required to lift the liquid the height of the inner diameter, against gravity, is just the

surface-tension-gradient force times the liquid flow velocity. When all this is combined the temperature gradient across the shell is directly proportional to the liquid layer thickness. Thus the ratio depends only on a combination of such parameters as the liquid and vapor densities, the heat of vaporization, etc., which determine the temperature dependence. Most significantly, however, the ratio plotted in Fig. 5-10 should not depend on the shell diameter, so data from many experiments should fall on the same curve, with one proviso. The correlation should still be sensitive to the shell thickness and thermal conductivity. The interior fluid mechanics can only depend on the heat flow through the wall and the temperature distribution on the inside of the wall. If these are fixed, the levitated layer thickness is fixed and will not vary if the shell wall thickness is made an order of magnitude thicker and the temperature drop across the shell wall is, therefore, a factor of ten greater.

For further information, please contact Dr. D. Bittner (WJSA).

5.3.3. SYMMETRIZATION OF SOLID FUEL LAYERS

This work has been carried out by Dr. Cho-ming Chen, a postdoctoral fellow at UR/LLE, under the supervision of UR/LLE. He had earlier demonstrated the possibilities of his approach by symmetrizing Ne ice at ILE, Osaka University.

On arrival at UR/LLE, his objective was to demonstrate layer symmetrization with hydrogen ice. There were two steps: (1) design of a cavity which would produce a spherically uniform plasma in a shell, and (2) construction and operation of a cryogenic cavity to demonstrate hydrogen ice symmetrization.

A series of problems have delayed his project, and it appears now that Dr. Cheng will leave in February 1995 having designed and constructed the cryogenic cavity, but without operating it.

Uniform Plasma Demonstration. A room temperature cavity was constructed to verify a design which would yield a uniform plasma inside a 3 mm diameter quartz sphere. In August 1994 a plasma was created in this sphere using 50 W at 3 GHz (25 μ s pulse width, 2% duty cycle). The shell was filled with a D₂ gas pressure of $\approx$ 20 Torr. There is a short time delay to create a discharge after the power is applied to the cavity; apparently an ionization event (cosmic ray?) is necessary to start the plasma.

Using Paschen's law (relating gas breakdown voltage to container dimensions) and the previous data acquired in Japan, the highest pressure at which a plasma can be ignited is 66 Torr. A glow discharge can be achieved at 20 Torr, which allows a significant pressure range over which to explore the relationship between the breakdown potential required to ignite a plasma and the shell gas pressure.

Preliminary infrared (IR) camera measurements of the shell temperature show a uniform heating effect from the plasma. Measurements are continuing.

Ice Layer Symmetrization. A cryogenic cavity has been designed and has been sent out for bid. To reduce the volume, I consists of a sapphire block containing a spherical cavity. The optical ports of the cavity are curved to reduce the strong lensing caused by sapphire's high index of refraction; this also helps with field uniformity. To confirm the cryogenic design, machineable high dielectric constant (HI-K) rod will be put into the modified cryogenic cavity to study the shift of the resonant frequency. After that the sapphire resonant cavity for the cryogenic experiment will be built. The D₂ fuel redistribution experiment will be done after the cryostat is built.

The room temperature experiment for estimating the heat generated from the plasma and the uniformity of the heating will be performed in the near future. These data will be the basis for knowing the operating range under cryogenic temperature conditions. The relationship of the variables (D₂ gas pressure, pulse width, duty cycle, and input power) will be investigated. It will not be possible to finish these experiments since the support of Dr. Chen ends and he must leave in February.

For further information, please contact Dr. R. Stephens (GA) or Dr. D. Bittner (WJSA).

5.3.4. FUTURE DIRECTIONS IN CRYOGENIC LAYERING

We are in the process of performing experiments with plasma assisted beta-layering and next generation thermal plate and laser heating experiments.

Plasma Assisted Beta-layering (Joule Heating). An extensive experimental program studying beta-layering in tritiated ICF targets has been under way for several years [Refs. 5-16,5-17]. Beta-layering is driven by internal heating of the solid due to the beta-decay of tritium. Inhomogeneities in layer thickness cause local temperature variations,

which result in material moving from warm to cold spots until an equilibrium isothermal surface is obtained. Unfortunately, the best estimates to date for a beta-layered surface has a roughness of 2 μm rms [Ref. 5-18]. This layer does not yet meet the surface finish specifications for a direct drive target for Omega Upgrade.

LLNL has done work looking at the surface roughness of non-tritiated hydrogen layers [Ref. 5-19] and found that by increasing the heat flux through the surface, smoothness is greatly improved. This process differs from beta-layering in that the gas is directly heated, and the gas then heats the solid by conduction. Thicker regions become warmer and, just as in beta-layering, material moves from warmer regions areas to cooler regions.

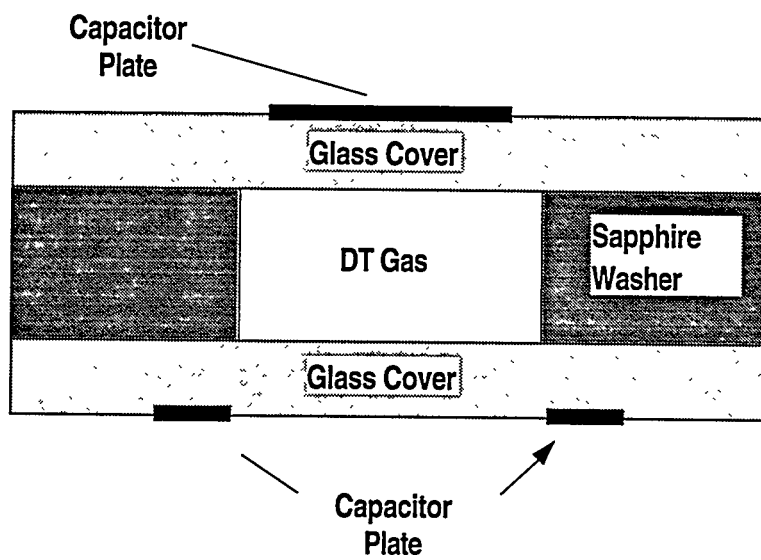
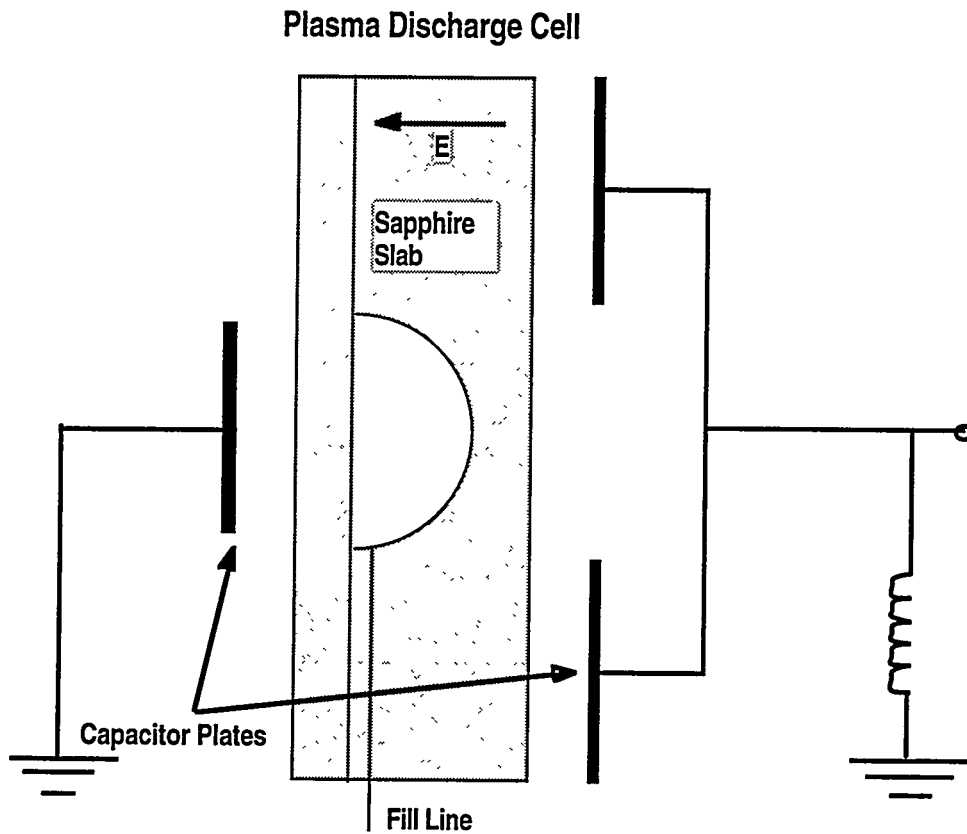
We plan to replicate the smooth surfaces obtained in the non-tritiated system in a shell filled with D-T. We will assist the beta-layering process by accelerating free ions in the gas with a RF field. The free ions are created by collisions of beta-particles with gas molecules. These accelerated ions then heat the rest of the gas by collisional transfer of energy. There is no dielectric breakdown within the gas, which differentiates this heating mechanism from plasma discharge heating.

The design of the experimental cell was begun this year. The cell will consist of an RF cavity (essentially a capacitor) which will enclose a glass or sapphire hemi-shell containing D-T. The temperature of the D-T will be maintained below the triple point but warm enough to maintain a sufficient vapor pressure. The cavity will be part of a resonant circuit driven by a standard RF source.

Figure 5-11 is a schematic representation of the initial design. This cell has the advantage of simplicity and can be cooled by anchoring the edge of the sapphire with a copper support.

Another cell design which will emulate the cylindrical cells that J. Hoffer and J. Simpson use at LLNL is shown in Fig. 5-12. Both cell designs have gaps to allow visual observation of the D-T surface.

It is important to know the shape of the **E** field inside the different cell designs. The complexity of the interior dielectric shape and of the plate designs compelled us to perform numerical calculations. Figure 5-13 shows the calculated **E** field for a cell which is similar to Fig. 5-11.



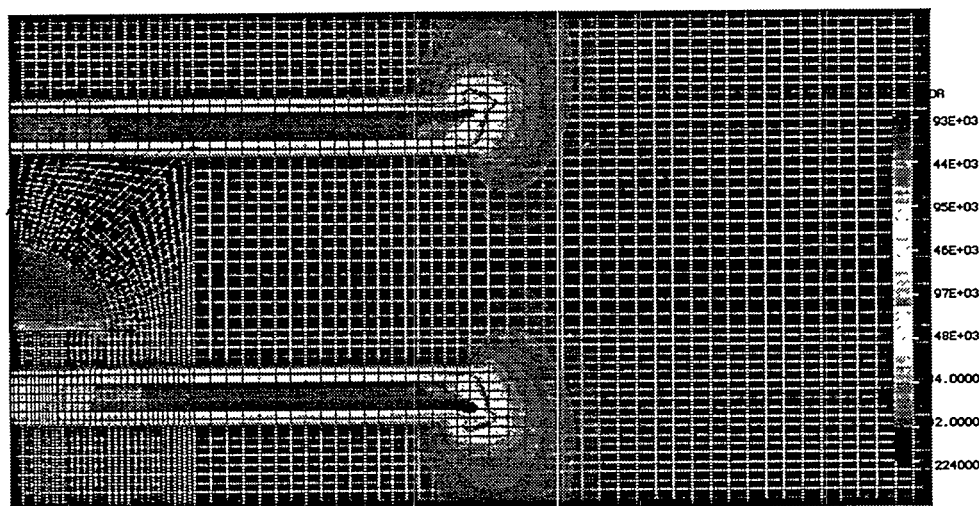


Fig. 5-13. An E field map (V/cm) for the experimental cell in Fig. 11. Only half of the cell is shown. Cylindrical symmetry is assumed.

The analysis was done for a static electric field with a potential difference of 400 volts applied to the capacitor plates. The plates are 1.8 mm apart and 10 mm in diameter with a 0.3 mm vacuum gap between the sapphire slab and the plates. The hemispherical cavity has a 1 mm radius. Most of the potential drop occurs in the gap between the plates and the sapphire. This effect can be reduced by decreasing the gap. Perhaps a set of capacitor plates can be vapor deposited directly on the sapphire. This second solution has the drawback of dumping energy dissipated in the plates directly into our sample. The benefit is a higher E field in the hemisphere for a given input power.

Square Cell Experiments. A significant fraction of time was spent designing and assembling cryogenically leak tight cells for use in the thermal plate and laser heating experiments. Figure 5-14 shows three square cell designs. Figure 5-14(a) is a sketch of the thermal plate experiment. Figure 5-14(b) is a sketch of the laser heating cell. The basic difference between the two cells, in addition to laser heating cell being ~25 mm longer than the thermal plate cell, is that the top copper endcap has a prism mounted on it for directing the laser light. As a backup design [Fig. 5-14(c)], if there are more problems with the infrasil glass, the infrasil tube in the laser heating experiment is replaced by a piece of invar with five infrasil windows.

These cells in Fig. 5-14 are for a next generation series of experiments, which will be performed using ~1 mm shells with ~40 μm thick wall made from free standing plasma polymer layers. The goal for these experiments is to demonstrate uniform layer

generation. An advantage with this experimental design is the helium exchange gas cooling is constrained to the region immediately surrounding the capsule. This will avoid a problem observed in previous experiments where the helium exchange gas filled the entire cold space and limited the amount of power which could be put across the shell without raising the overall temperature of the experiment.

In testing the new cell designs a few problems which resulted in loss of cell integrity were encountered. After making a few changes, we have now repeatedly made and broken the indium seals and cycled to liquid nitrogen temperatures with retention of cell integrity. We are currently in the process of mounting capsules and preparing two cells each for the thermal plate and laser heating experiments.

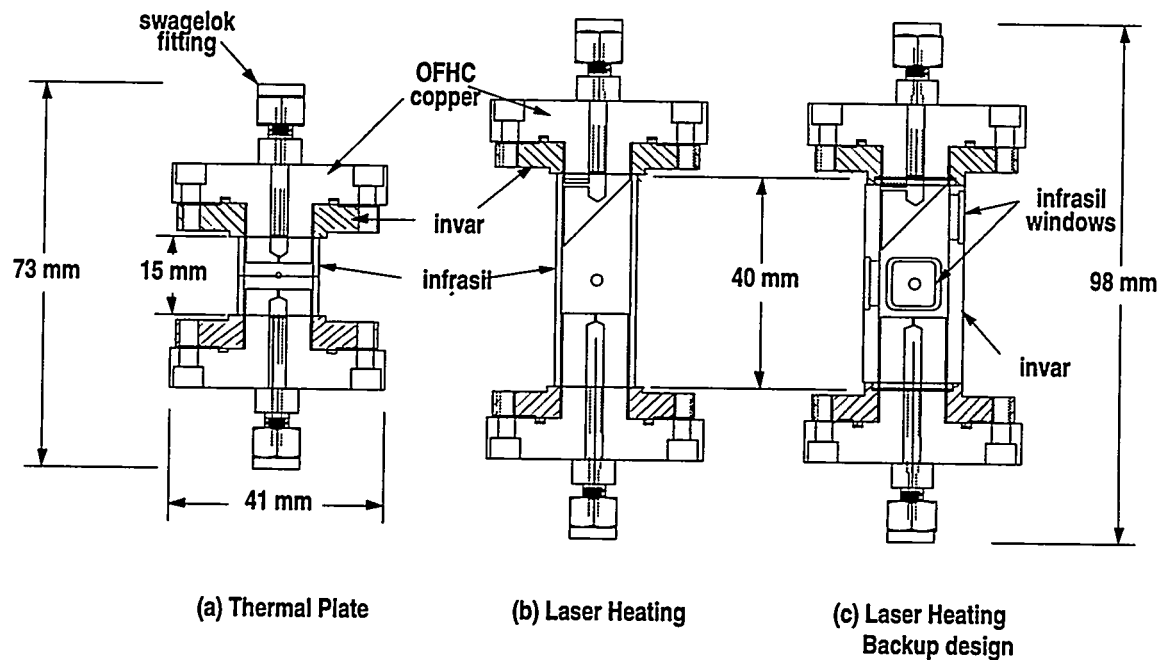


Fig. 5-14. Designs for the square cell experimental series.

For further information, please contact Dr. D. Bittner (WJSA).

5.4. CRYOGENIC LAYER CHARACTERIZATION DEVELOPMENT

The objective of this task is to develop a technique to characterize the roughness of the inner surface of a shell. We proceeded in two parts: 1) to develop a measurement technique, and 2) to relate the measurements to the inner surface roughness.

5.4.1. MEASUREMENT TECHNIQUE

During the course of this year, we have found a potentially acceptable optical profiler and have developed a conceptual protocol for using it to extract buried surface L-mode information. In the next year we expect to do analyses to finalize an instrument configuration and to determine its measurement limitations.

Chapman Instruments makes an optical profiler which has demonstrated the capacity to characterize the bottom surface of a thin plastic sheet when it has sufficient magnification (50× or 80×). It can measure a line of spots 1 mm long, on surfaces tilted up to 21° from horizontal. It measures the spots sequentially, at a rate of 2,000/s. It actually measures the slope at each spot, by comparing the optical path difference between two adjacent (0.2 μm separation), perpendicularly polarized spots.

One could make an optical analog of LLNL's AFM Sphere Mapper using this head. It's lateral translation capability would allow characterization of a large part of the shell surface ($\approx 35\%$) in a single measurement. The optical head would collect data on a zig-zag set of lines extending $\approx 400 \mu\text{m}$ North and South of the equator, while the sphere (1 mm diam.) rotated underneath it. If it collects at 1 μm intervals, each line ($\approx 800 \mu\text{m}$ long) would take 0.4 seconds to characterize, and would contain information about modes 10 to 6,000. If the shell is rotating at a rate of once per 40 seconds, 100 such lines are collected, which can be combined to give information about modes 2 to 100.

Other measurement protocols are possible; measurement along a set of independent great circles is one. The scheme described above has the advantage of minimizing shell handling.

5.4.2. INNER SURFACE CHARACTERIZATION

Analysis of buried surface characterization can proceed in two ways: analysis and modeling.

Inner Surface Measurement Analysis. One could consider the upper surface ripples to be lenses which modify the apparent focal plane of the microscope. That modification imprints some of the upper surface roughness on the measured lower surface. If it is assumed that the roughness perturbations are small enough that effects are linear, then this is susceptible to a rather simple analysis.

Setup:

Image the surface through an objective with a given focal length (f) and numerical aperture (NA). The numerical aperture of the lens, $NA \equiv \text{lens-radius}/f$.

The surface being observed is the bottom of a layer of some thickness (th) and index of refraction (n). Assumes observation is from directly overhead.

The top surface of the layer has ripples on it with some wavelength (λ) and amplitude (amp).

Effects:

The apparent surface height of the inner surface is modified by $amp \sin(2\pi r/\lambda)$, where r is the distance along the ripple.

The overlayer ripples also cause an uncertainty in the angle of the upper surface of $\theta_1 \pm 2\pi amp/\lambda$.

Snell's law gives $\sin \theta_1 = n \sin \theta_2$ which for small angles is $\theta_1 = n\theta_2$. The change in angle, $\Delta\theta = (\theta_1 - \theta_2)$ of a beam incident at θ_1 is $\Delta\theta = (1 - 1/n) \theta_1$.

This gives fluctuations in apparent lower layer location of $th \times \Delta\theta$. Maximum $\theta_1 = NA$.

If the observation rays go through the upper surface along a length (b) longer than λ , the wavelength of the upper surface ripple, the image of the lower surface is laterally averaged over length $2th\Delta\theta$. If b is much shorter than λ , its lateral position is merely distorted by up to $\pm th\Delta\theta$.

The apparent height of the inner surface is changed by the ripple because of the focal length of the lens it creates. For a flat layer, the upper surface appears at depth th/n . The radius of curvature (rad) of a sinusoid varies between $\pm(\lambda/2\pi)^2 \times 1/amp$. It forms a

lens with focal length (foc) varying between $\pm \frac{n_1}{(n_1-1)}$ rad. This modifies the focal length of the objective by the sum of their reciprocal focal lengths. As a result, the height of the lower surface is modified by $\frac{1}{(1/f \pm 1/\text{foc})} - f$. In the case of short surface ripples, the height is fuzzed out, in the case of long ripples, it is distorted.

These considerations will give the following:

Short wavelength ripples (short defined by objective NA and layer thickness) fuzz out lateral and vertical information by calculable amounts, and put a lower limit on amplitude, and an upper limit on fluctuation wavelength which can be observed in the lower surface. The limits are determined by some kind of average of amplitude over wavelength of short wavelength ripples.

Long wavelength ripples distort both laterally and vertically and add apparent ripples to real distortions of the lower surface. This is a lower limit in amplitude vs. wavelength which is the upper surface spectrum times a transfer function which goes to zero for very long wavelengths and goes to zero again as the wavelengths approach the short regime. The maximum effect occurs at a wavelength about equal to b .

Inner Surface Measurement Modeling. Modeling the optical setup is more tedious, but can include realistic effects, and can be used to establish the amount of data collected by a specific protocol in fact required to confidently characterize a surface.

Pat McKenty (UR/LLE) has been working with computer generated sphere surfaces, and has found, for instance, that using the data from nine great circles is sufficient to characterize a sphere surface.

Rich Stephens (GA) is starting by modeling the behavior of the Chapman Instruments head. Its double beam system measures the difference in optical path-length between reflections from adjacent spots on an interface; it is not trying to focus on a surface. That can be modeled by calculating the optical path length for reflections off a buried surface. That path length will determine the apparent position of the buried surface. Rough flat surfaces have been generated with specified power spectra, and a program is being written to calculate the apparent roughness of a smooth buried surface. Eventually this program will be made more sophisticated; it will generate and work on spherical surfaces, and consider the added roughness of an already rough buried surface.

For further information, please contact Dr. R. Stephens (GA).

5.5. FOAM SHELLS FOR CRYOGENIC TARGETS

The objective of this task was to make hollow foam shells, with a full density polymer overcoat to the following specifications:

Outside Diameter = 700 to 1400 μm

Overcoat Thickness = 5 to 15 μm

Foam Thickness = 50 to 130 μm

Foam density <50 mg/cm^3

A foam shell technology has been developed for this purpose at ILE at Osaka Japan, the lead investigators being Norimatsu and Takagi [Refs. 5-20, 5-21]. (See Fig. 5-15 for an overview of the process.) However, the typical Osaka shell is 500 μm in diameter with a 10 μm wall. Our goal was to determine whether this technology could be successfully adapted to meet the target requirements of the Omega Upgrade and the capsule requirements of the National Cryogenic Program.

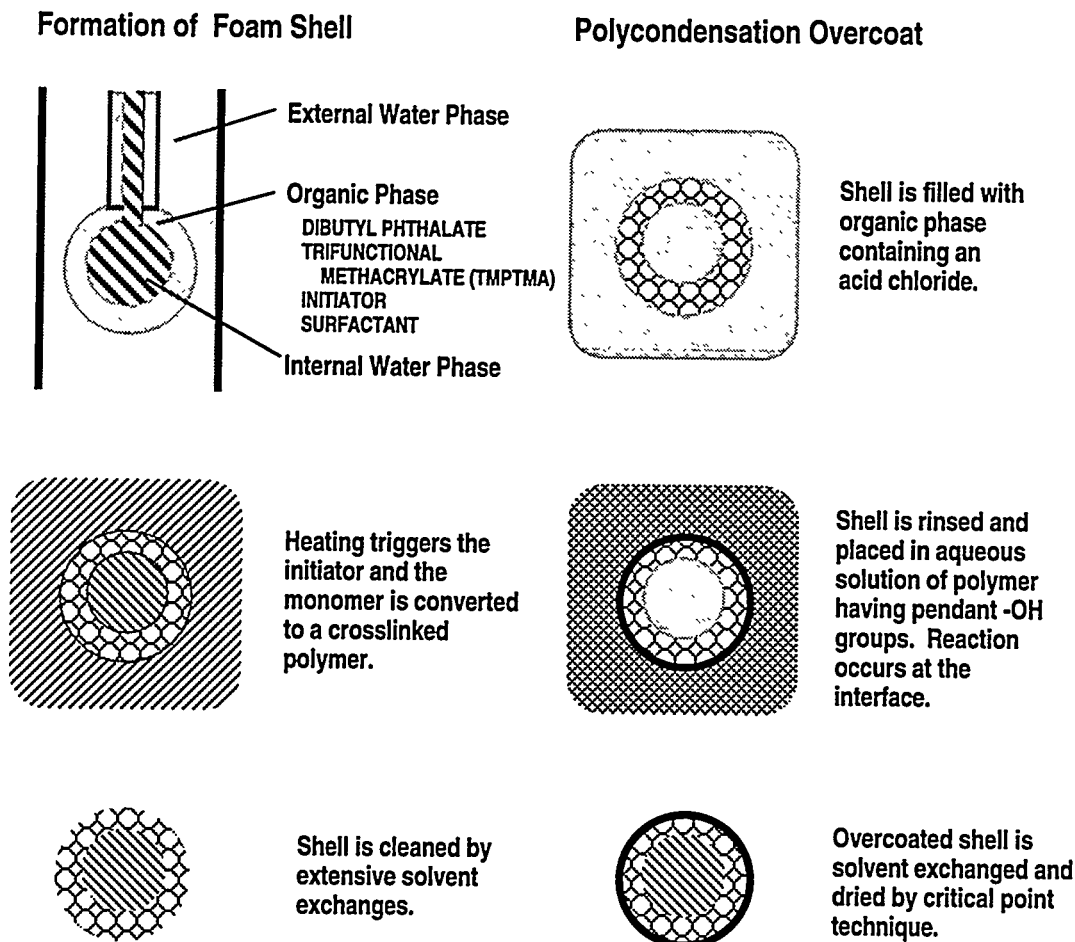


Fig. 5-15. A pictorial representation of the foam shell process.

5.5.1. FOAM SHELL PRODUCTION

The production of foam shells began at the General Atomics facility, using a duplication of the Osaka "Shake and Toss" technique. Shortly thereafter, the task was changed to an "onsite" task to combine the contract task with an ongoing LLNL task. With this transfer the production method also changed; a droplet generator was developed. The generator was modeled after an Osaka drawing of their generator, but no frequency modulation was used. For shells 1 mm in diameter, none was needed.

The droplet generator produced shell preforms of the desired diameters and walls thicknesses, but gelation of the preforms was inconsistent. Random batches of shells would dissolve when exposed to a toluene exchange. It appeared that the preforms were undergoing some degree of polymerization (due the observed shell cloudiness), but that the polymer was not crosslinking. To understand the factors effecting polymerization, a series of viscosity studies were undertaken. Bulk samples of the organic phase were heated in a Rheometrics viscometer, to induce polymerization, and the viscosity increase as a function of time was measured.

Two routes were explored to facilitate the polymerization process: increasing the concentration of the radical initiator AIBN (2,2-azobisisobutyronitrile), and removal of oxygen. Actually these two approaches are linked since removing oxygen from the system has the effect of increasing the concentration of available initiator. Oxygen is an inhibitor which reacts with propagating radicals to form a relatively unreactive peroxy radical. This radical is able to react with either itself or another propagating radical, but this results in chain termination.

The concentration of initiator was varied from its historic value of 0.032M (1x in this context) to two times (2x) and to three times (3x) for both degassed (i.e., oxygen removed) and untreated (i.e., "not degassed") samples. Figure 5-16 shows the sample viscosity as a function of time at 60°C for each of the six samples. It is proposed that the high inhibition constant for oxygen explains the difference in time to the onset of gelation between the degassed and not degassed samples for the 1x or 2x initiator concentrations. The fact 3x degassed, 3x not degassed and 2x degassed all have the same gelation onset time suggests there is an induction period for the gelation of TMPTMA in dibutyl phthalate. This may be due to the necessity of having a minimum chain length before entanglement and crosslinking can effectively take place. The time to reach this chain

length would be governed largely by monomer concentration rather than initiator concentration. Also, the qualitative shape of the curves suggest that increasing initiator concentration decreases the time from the start of gelation to the final gelled state. Based upon these results, we adopted a standard procedure of using the 2x initiator concentration and degassing the organic phase and both water phases.

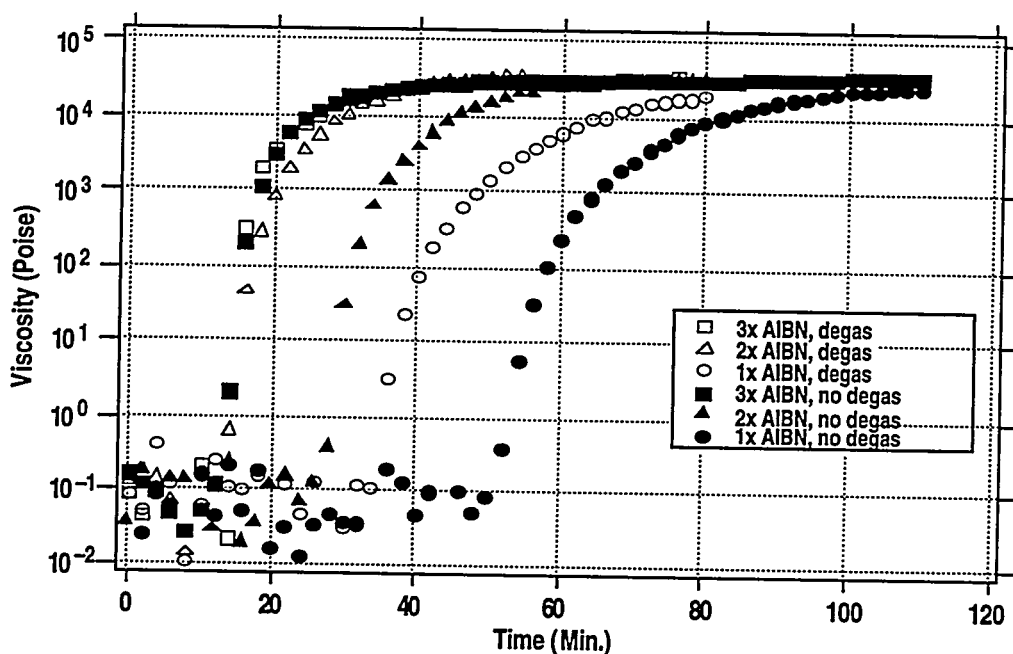


Fig. 5-16. This graph depicts the results of experiments undertaken to decrease the time to gelation. The concentration of the initiator and removing exposure to oxygen were key.

The initial foam shells produced had a narrow size distribution, but often severe non-concentricity. This non-concentricity was not a function of the production technique, but rather a density mismatch between the organic phase and the aqueous phases. The organic phase was primarily dibutyl phthalate which has a density of 1.05. Thus the organic phase would sag around the more buoyant internal water phase. Toluene was selected as a compatible, lower density solvent to blend into the organic phase to reduce its density. Calculations indicated a 12% toluene and 88% dibutyl phthalate solution would be density matched at the polymerization temperature, 90°C. The shells produced with the solvent blend have significantly improved concentricity. The average non-concentricity (difference of two walls divided by the average wall) has been as low as 5% for shell batches and less than 1% for individual shells. Figure 5-17 is a photo of unsorted, 1 mm shells with 100 mm walls, produced by the blended solvent system in the droplet generator.

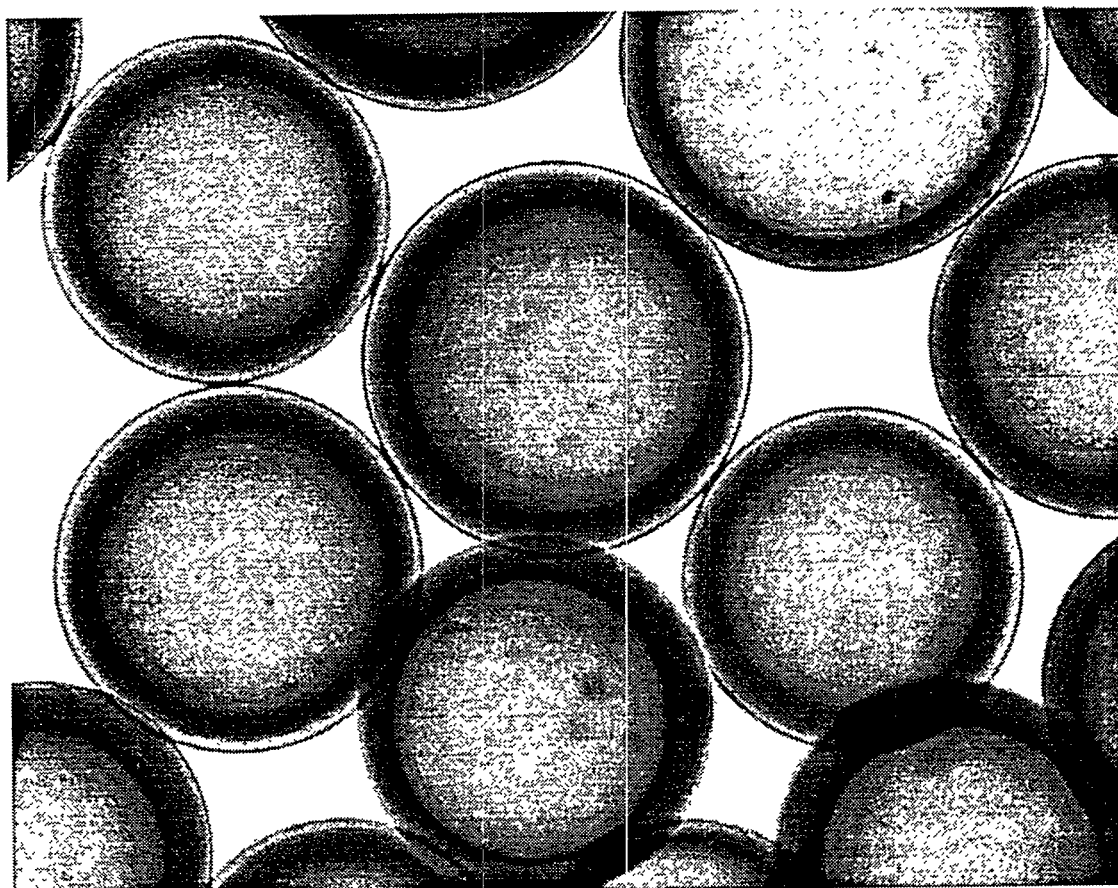


Fig. 5-17. A typical shell batch, 1 mm in diameter with 100 nm walls. The shells are immersed in ethanol for better transparency.

We overcoated the foam shells first with hydroxy ethyl cellulose. The reaction easily produced overcoated shells, but the surface finish was poor. The recent Osaka literature indicated that using poly(vinyl phenol) (PVP) produced superior surface finish, on the order of 0.2 micron roughness. Thus we switched to the PVP reaction. We were able to produce reasonable background surface finishes (less than 0.1 micron RMS as measured by AFM), but debris one micron in height was also observed. We have possible solutions to this problem, for example doing the coating in a clean area, but there was not sufficient time to explore them.

5.5.2. OPACITY ISSUES

The opacity of the shells produced was not listed in the task specifications, yet it is critical for their use by National Cryogenic Program. For liquid layer uniformity studies, the optical properties of the shell must be sufficient to allow for characterization of a

layer edge to within a micron. The opacity of the shell is due to the relatively large cell size of the foam structure ($1\text{--}2\text{ }\mu\text{m}$) coupled with an index of refraction mismatch between the foam ($n = 1.55$) and liquid hydrogen ($n = 1.13$). To characterize the optical transparency as a function of foam thickness, foam wedges were cast. Three foam wedges (filled with air, $n = 1.0$) were measured for percent transmission using a Leitz Orthoplan microscope and photometer at $\lambda = 641\text{ nm}$. The results are displayed as open symbols in Fig. 5-18. Two of the wedges were then filled with ethanol ($n = 1.32$) and perfluorohexane ($n = 1.25$) and measured again. Based upon this crude analysis it appears that we can expect less than 10% transmission through 200 mm of liquid hydrogen filled foam. This will not be adequate for liquid layering studies given the current optical diagnostics being used.

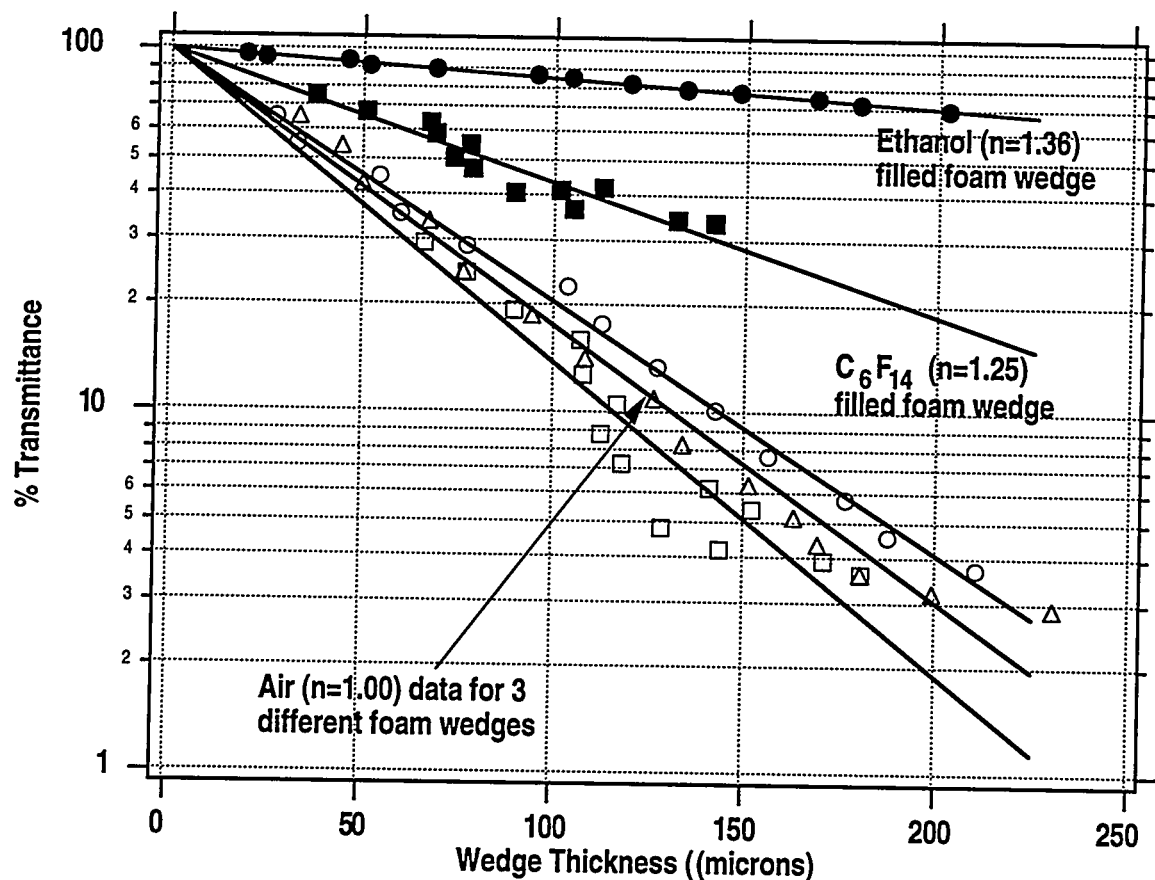


Fig. 5-18. This graph indicates the percent of optical transmission as a function of foam thickness. The transmission is a function of both the foam and the index of refraction of the liquid that fills it.

5.5.3. CONCLUSION

Based upon the work reported, we believe that microencapsulation is a viable technique to meeting the foam shell needs of the ICF program. Foam shells meeting task specifications were produced by adapting the methods and chemistries developed by ILE. This was made possible by gelation studies which furthered understanding of the factors controlling polymerization.

However, we cannot state that the current optical characteristics will meet the diagnostic requirements of the National Cryogenic Program. The current product with 100 mm walls has an optical transmission of less than 10%. This will not allow the characterization of the liquid layer to within a micron. Therefore, experiments will be done in FY95 to reduce the TMPTMA foam cell size (as recently reported by ILE) [Ref. 5-22] and to apply the microencapsulation technique to a transparent aerogel, resorcinol-formaldehyde. Both approaches will be explored until one technique is proven able to meet both the dimensional and optical specifications.

5.5.4. ACKNOWLEDGMENT

We are pleased to acknowledge the cooperation of the researchers of ILE, especially Dr. Norimatsu and Dr. Takagi.

For further information, please contact D. Schroen-Carey (WJSA).

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SECTION 6

CRYOGENIC SYSTEM DESIGN FOR OMEGA UPGRADE

6. CRYOGENIC SYSTEM DESIGN FOR OMEGA UPGRADE

The upgrade of the OMEGA laser at the University of Rochester's Laboratory for Laser Energetics (LLE) to 30 kJ (351 nm) will result in a need for millimeter-size Inertial Confinement Fusion (ICF) targets filled with D₂ or D-T and maintained at cryogenic temperatures. This mandates a cryogenic target delivery system capable of filling, layering, characterizing and delivering cryogenic targets to the OMEGA Upgrade target chamber. General Atomics has been assigned the task of designing, constructing and testing the cryogenic target delivery system.

6.1. FILL STATION ACTIVITIES

The D-T fill station at UR/LLE was designed to fill glass targets at elevated temperatures for the OMEGA experiment.

6.1.1. MODIFICATION OF UR/LLE FILL STATION

Several modifications to the existing fill station were required to fill low pressure gas targets for OMEGA Upgrade. The modifications included adding an annex to the existing glovebox to house target characterization equipment. Other changes included replacing the existing open cycle liquid helium system with a closed cycle cryocooler and redesigning the contact tube to allow use of the GA designed target rack and redesign of the condensation tube to minimize system dead volume.

Modifications were also required to allow for simple attachment of the high pressure D-T permeation cryostat fill system. Once the fill station is in operation, modifications will be difficult unless provisions are made before the system is charged with D-T. An example of this type of modification is the addition of connections to the D-T plumbing. Other examples include relocating the existing fill station to provide space for the high pressure system and adding utilities such as an overhead crane and liquid nitrogen plumbing.

For further information, please contact C. Gibson (GA).

6.1.2. FILL STATION

The targets are placed in a permeation cryostat (Fig. 6-1). The D_2 fill station will use an existing D_2 fill station to reach 150 atm pressures (at room temperature). A high pressure gas intensifier, acting as a second stage, increases the working pressure in the permeation cell (Fig 6-2. and item ① in Fig 6-1) to 1100 atm. To keep the shells from collapsing during pressurization, the thin walled shells are filled in steps no greater than the degree dictated by the strength of the shells (<10 atm). A stepper motor driven gas intensifier was selected because of the need for high accuracy pressurization steps (0.2 atm/step). After the permeation fill, the targets are cooled below the triple point of D_2 or D-T (reducing the internal pressure to a few atm to prevent the shells from bursting). The cryogenic valves ② are opened and the excess fuel (outside the target) is pumped away ④ by a turbomolecular drag pump. A cold transfer cryostat is used to remove the targets from the permeation cryostat.

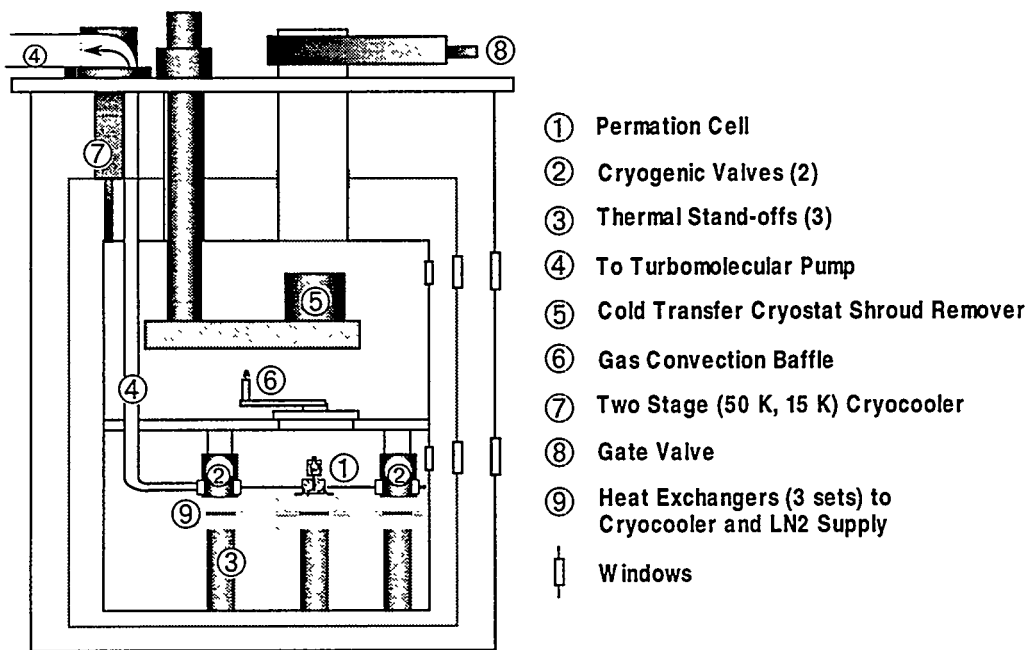


Fig. 6-1. Permeation cryostat showing major components.

Activities for FY94 included repair and modification of the high pressure intensifier. After purchase, the intensifier developed a slight leak which required repair. Additionally, Teflon and Vespel packing rings were used for sealing the intensifier. Because the intensifier will ultimately be used for tritium handling, elimination of the

Teflon packing was mandatory. The chrome plated stainless steel intensifier piston was replaced with a polished tungsten carbide piston, and the packing was replaced with a pure Vespel seal ring. The intensifier is effectively leak free ($<1/3$ psia/day, at 20,000 psia) after 500 cycles.

The high pressure cryogenic components for the prototype D₂ fill station were designed in FY94. These include the high pressure target permeation cell, the high pressure cryogenic valves, and the cryogenic pressure transducer. At UR/LLE request, the permeation cell (Fig 6-2.) was modified to accommodate longer target standoffs, and the target rack modified to include fratricide shields.

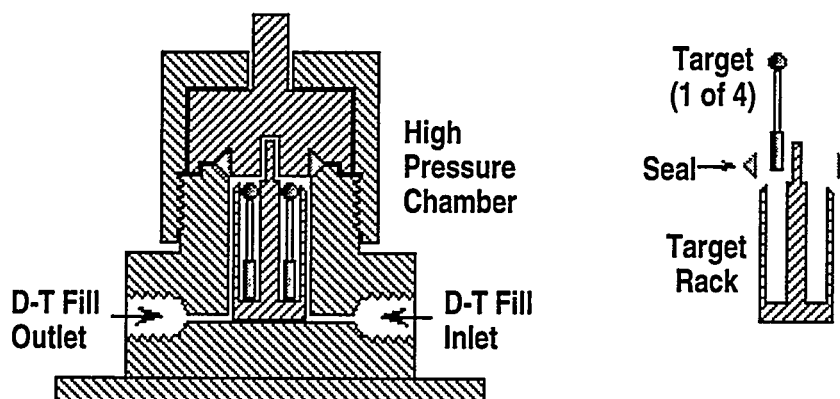


Fig. 6-2. Permeation cell.

These drawings were released and are currently being fabricated. The high pressure cryogenic valves were designed and are being built. The high pressure (20,000 psia) cryogenic transducer has been tested to LN₂ temperatures and physically survived. Low temperature electronics are required for 20 K operation.

For further information, please contact R. Mangano (GA).

6.2. CRYOGENIC TRANSPORT

We have implemented a change in the baseline conceptual design that will significantly improve performance. The new concept for moving filled targets from the fill station's permeation cryostat to the layering and characterization cryostat utilizes a single cold transport cryostat. A vacuum bellows manipulator is used as the outer vacuum container of the cold transport cryostat. Through a pair of gate-valves forming a vacuum lock, the cold transport cryostat attaches to the top of the roof of the glove-box. This lock is directly connected to the neck of the permeation cryostat, which is inside the permeation cryostat. Opening the gate-valves and collapsing the bellows drives the cold inner portion of the cold transfer cryostat into the bottom of the permeation cryostat to retrieve filled targets from the permeation cell. After the targets are seized the bellows are extended and the vacuum lock closed.

The cold transfer cryostat is then detached from the glove-box. The cold transfer cryostat is transported with a mobile power cart to the layering and characterization cryostat and a similar process is used to deposit the targets into the layer and characterization cryostat. Mobile power is required for the cold transfer cryostat's closed-cycle, two stage, Gifford-McMahon cryocooler. This contrasts with the prior concept, which used an intermediary storage dewar for transport between the filling and layering stations. In the old concept, one cold transfer cryostat inside a large glove-box transferred the targets from the permeation cryostat to the storage dewar. The storage dewar passes through a large vacuum lock to arrive outside the glove-box. The storage dewar is transported next to the layering and characterization cryostat, where a second cold transfer cryostat is used to transfer the targets into the layering and characterization cryostat. Figure 6-3. illustrates these concepts. Both concepts use a low thermal conductivity cryogenic extension wrench to open the permeation cell, inside the permeation cryostat, before the cold transfer cryostat is used to retrieve the targets. In the new concept, a vacuum bellows manipulator is provided for using the cryogenic wrench in the same manner as it is used with the cold transfer cryostat.

This new concept is safer. The operating personnel do not have to handle surfaces that have been exposed to the glove-box atmosphere. There are half as many cold transfer

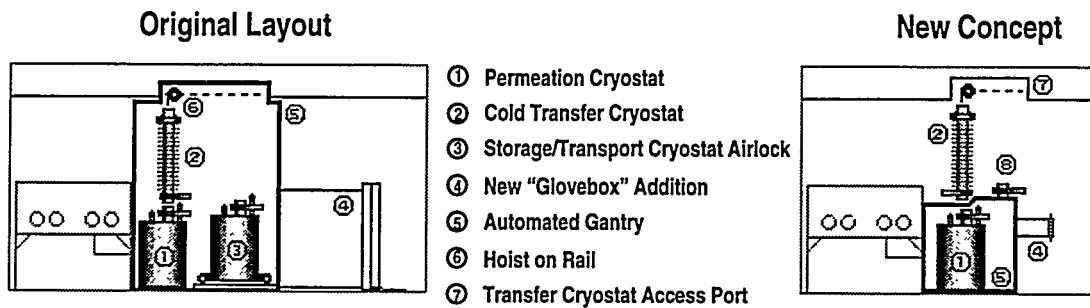


Fig. 6-3. Illustration of cryostat transfer.

operations per layered target which translates into lower probability of accident. The new concept is less expensive. One cold transfer cryostat and the storage dewar have been eliminated. Target inventory is now kept in the layering and characterization cryostat. The glove-box size is smaller and it no longer contains an automated gantry for maneuvering the cold transfer cryostat. The new concept is faster. There are half as many cold transfer operations per layered target which translates into reduced cycle time. Moving operation and maintenance of the cold transfer cryostat outside the glove-box also speeds operation.

Successful functioning of the cold transfer cryostat (Fig. 6-4) requires a vacuum seal that can be resealed at cryogenic temperatures (~ 20 K) at least once and preferably

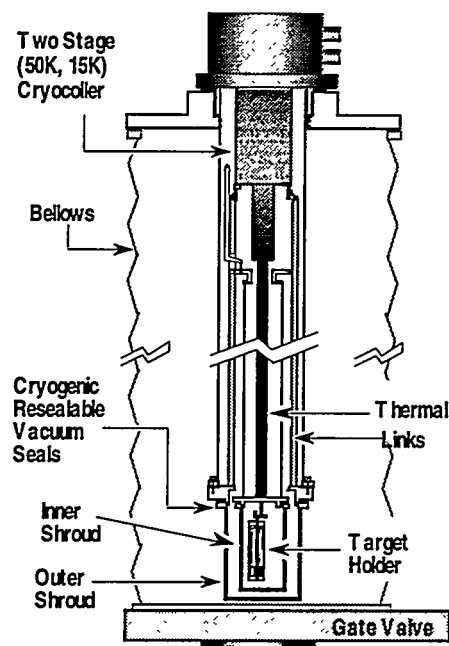


Fig. 6-4. Cold transfer cryostat.

many times. Using the cryogenic vacuum seal testing apparatus built last year, we tested all the prospective seals mentioned in last year's annual report and several other designs (including Teflon®, OFHC copper, indium plated OFHC copper, platinum, stainless steel, silver plated stainless steel, nickel, and silver plated nickel). The best performing seal designs (in terms of low leak rate and multiple low temperature reseals) used 316 stainless steel flanges with hemispherical ridges pressing into indium plated OFHC copper gaskets.

Figure 6-5 shows the test results for these seals. Seal A was made from OFHC Copper gasket plated with 0.002 in. indium, soldered with indium to one flange. The other flange was a hemispherical ridge. Seal B was made from an OFHC Copper gasket plated with ~0.00025 in. indium on both flanges. The seals were opened and closed at 15 K with a differential helium pressure of 100 Torr. This level of performance will work very well in the cold transfer cryostat application.

All the systems needed to demonstrate the cold transfer of targets were designed in official drawings and/or specified in a technical specification, and ordered this year. The systems are as follows: the permeation cryostat; high pressure cryogenic valves,

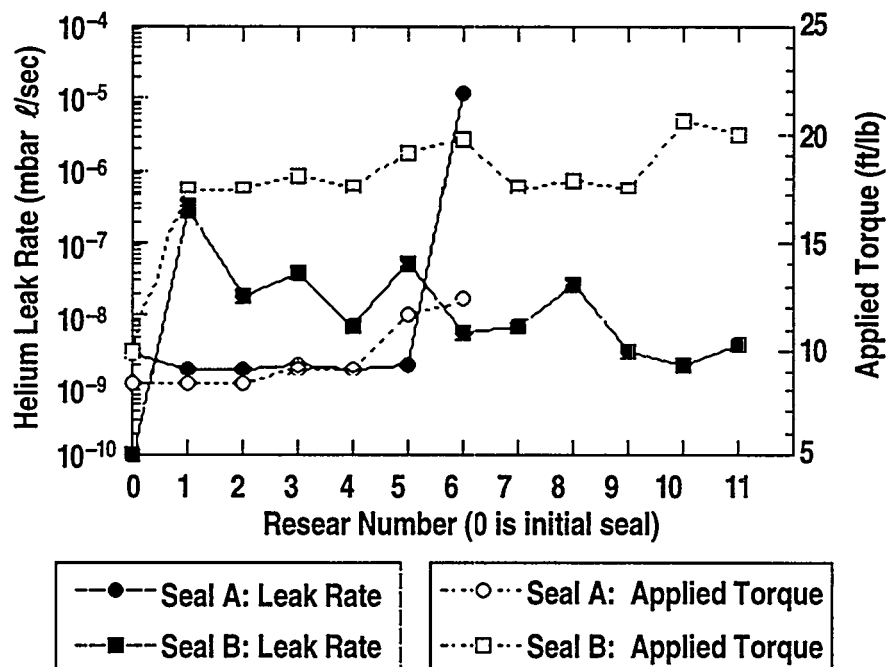


Fig. 6-5. Seal test results.

located inside the permeation cryostat; high pressure permeation cell, located inside the permeation cryostat; shroud manipulator, installed inside the permeation cryostat for use in opening and closing the cold transfer cryostat shrouds; cold transfer cryostat; vacuum bellows manipulators; cryogenic wrench; target base; target rack (used to hold multiple targets in the cell); and target manipulator used by the cold transfer cryostat. Major construction activity will take place in FY95 when the major subsystems are received from the vendors.

For further information, please contact N. Alexander (GA).

6.3. TARGET INSERTION

We explored several concepts for target insertion. Two concepts, the cryogenic elevator and the insertable cryostat, are still being actively pursued with a third, augmented mass free fall, remaining as a backup, primarily for liquid layered targets. The cryogenic elevator is based on the original design concept and the insertable cryostat was developed as response to problems in meeting the design requirements. Several of the design requirements were also investigated to translate goal oriented requirements into design specifications.

Other than the target support, the end of the fast shroud will be the closest material to tank center at the time of the shot. An investigation was made of the probable x-ray driven ablation of various materials to determine which materials could be permitted near chamber center at shot time. Carbon appears to be most resistant to x-ray damage and will be used at the tip of the fast shroud. A 20 cm standoff distance appears to be acceptable and the tip of the fast shroud will be withdrawn a minimum of 20 cm from chamber center at the time of the shot.

Based on the coefficient of thermal expansion and rupture strength of solid hydrogen, we have assumed that the temperature of solid layered targets must be maintained with ± 0.2 K from the time they are layered until they are shot. There is no experimental basis for confirming or rejecting this assumption.

The withdraw time for the fast shroud for solid targets can be determined from the allowed temperature rise and the absorption characteristics of the target. Previously,

based on a target emissivity (ϵ) of 1, the shroud withdrawal time was determined to be 20 ms. New calculations, based on the absorption spectra of a thick hydrogen layer in a thin polystyrene capsule, indicate that 100 ms are available for shroud withdrawal. Because opaque targets have not yet been ruled out, we have maintained 20 ms shroud withdrawal as the design basis.

These design requirements/specifications were used as a basis for shroud withdrawal investigations. Calculations were made of peak shroud velocity, acceleration, and acceleration forces based on the assumed 20 cm withdrawal in 20 ms ($\epsilon_{\text{target}} = 1$) and estimates of shroud mass and actual coil masses of commercial variable reluctance linear actuators. The results indicated that simple acceleration of the shroud away from the target was unacceptable on the basis of all three parameters. All three parameters may be shifted to the upper range of commercially realizable actuators, if the shroud initially extends well beyond the target. This extended distance is used to accelerate the shroud so that it is traveling rapidly when the end passes the target exposing it to background thermal radiation. A 35 cm initial acceleration distance appears to be a reasonable compromise. Using ramped acceleration to minimize mechanically induced shock, the shroud is accelerated to 10 m/s over 35 cm in 130 ms and the end of the shroud clears the target in an additional 20 ms. The shroud is stopped within the next 10 cm giving an overall shroud travel of 70 cm. The peak acceleration under this scenario is almost 15 g's. This means that the mass of the fast shroud must be minimized if the acceleration forces are to be realizable. As mentioned earlier, theoretical calculations indicate that ϵ_{target} may be much less than 1. If true, there will be a commensurate reduction in the accelerations and peak velocities required.

Both the cryogenic elevator and the insertable cryostat have been changed from the original concept in that both the slow (outer) and fast (inner) thermal shroud are now withdrawn up into the upper probe. The decision to withdraw the fast shroud upward is a direct result of the analysis of shroud acceleration. If the shroud is to be of minimum mass and thus minimum diameter, the 5 mm target mounting pedestal would have to be a minimum of 90 cm long (and probably more like 125 cm long for downward withdrawal). If the fast shroud is withdrawn upward the pedestal will be slightly more than 35 cm long. Trying to control the position of a target on the end of a 5 mm diameter, meter long rod will be very difficult. Moreover, the moving portion of the target insertion system is already almost too long to permit loading of the target in the basement (*La Cave*) region below the OMEGA Upgrade target chamber.

6.3.1. CRYOGENIC ELEVATOR

This concept is based on a uniformly cooled environmental chamber extending from the Layering and Characterizing Module, in *La Cave* below the target chamber, to the center of the target chamber. A means is provided to raise the target, 7.5 m, from *La Cave* and position it at tank center. Preliminary estimates of temperature stability, based on thermal expansion and rupture strength data of solid hydrogen, indicate that the temperature of the target must be kept within ± 0.2 K of the layering temperature (~ 18 K) while being transported and while waiting at chamber center. Much of the target insertion positioning mechanism must operate at cryogenic temperatures at a pressure on the order of 1 Torr.

The binary thermal linkage concept (Fig. 6-6) was developed to provide a means for maintaining very uniform temperatures over relatively large distances. Preliminary calculations indicated that a single cryocooler, connected to concentric 80 and 15 K heat sinks could maintain the interior of a 6 m elevator housing within a few millikelvin.

Insertable Cryostat. The insertable cryostat (Fig. 6-7) consists of a small cryostat cooled by a two stage cryocooler. The cryostat is raised from *La Cave*. The upper

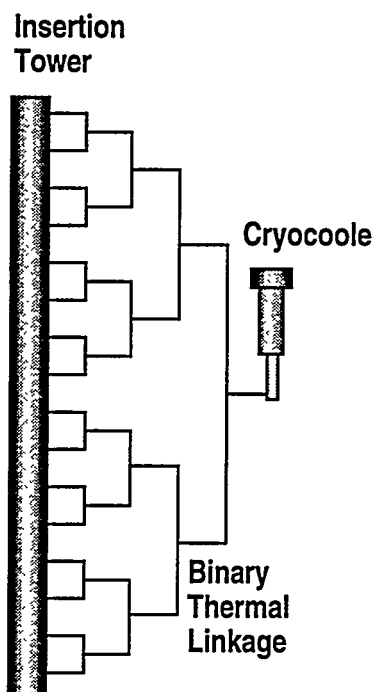


Fig. 6-6. Binary thermal linkage concept.

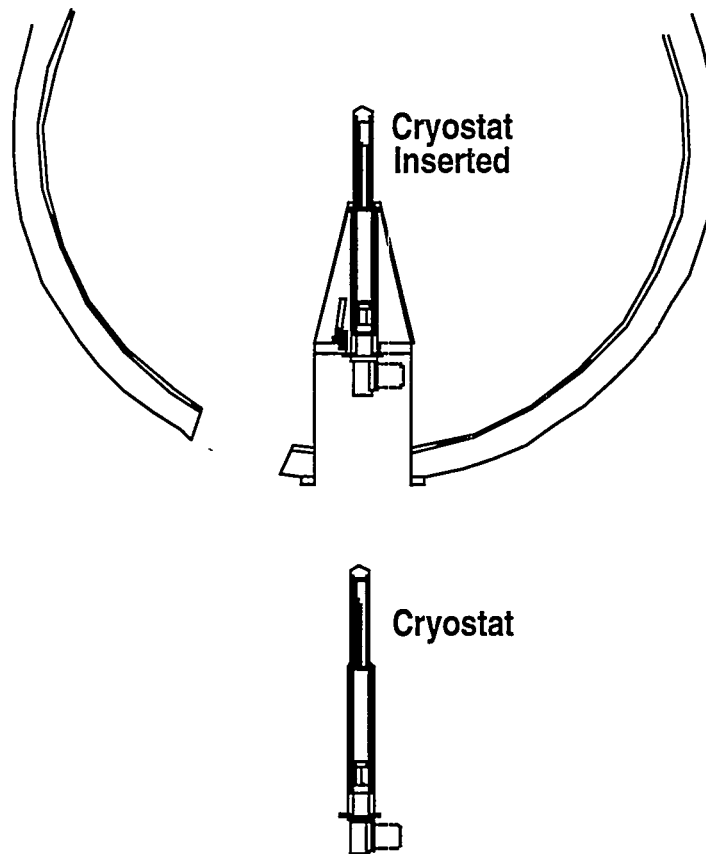


Fig. 6-7. Insertable Cryostat

end, containing the target, is inserted through an air lock into the center of the chamber. At the upper end of the cryostat is a pair of detachable shrouds which can be removed through a second probe extending from the top of the target chamber.

The insertable cryostat has many advantages over the cryogenic elevator but has its own design problems as well. Besides minimizing the volume to be maintained at cryogenic temperature and eliminating cryogenic operation of the insertion and positioning mechanism, this method would make investigation of alternative layering methods much easier. The layering and characterization apparatus may even be incorporated into the insertable cryostat. Two major design problems deal with the mechanism for disconnecting the shroud from the cryostat and designing the cryostat small enough to fit up through the port in the bottom of the chamber. The cryostat must fit up through the 18 in. port in the bottom of the chamber to the air lock location. If the air lock is not located up inside the chamber, the cryostat will be too tall to fit in the *La Cave* region below the chamber due to the extremely low head room (80 in.).

6.3.2. AUGMENTED MASS FREE FALL

Free fall has been considered both as a means of liquid layering and as a means of transporting the target to the center of the target chamber. Two factors have deflected sufficient accuracy and the second is the effect of residual gas drag on the falling target. Calculations on the London-Van der Waals forces [Ref. 6-1] indicate the target must fall with an acceleration within 1 ppm of local gravity to generate thin ($\sim 1 \mu\text{m}$) overfilled liquid layers. A 10^{-4} Torr residual gas pressure (maximum allowable pressure in the target chamber) will cause a drag of 10^{-3} g on a typical 1 mm diameter target. Because the drag forces are inversely proportional to mass, if the target is mounted to a larger mass, the relative effect of gas drag will be reduced. Simple calculations were made of the effect of mass augmentation (Fig. 6-8.). A mass equivalent to a 1 cm sphere of tungsten should have a sufficient mass/drag area that it falls with an acceleration within 1 ppm of true local gravity. The true effect of shape has yet to be assessed.

The augmented mass will also increase the accuracy of target aiming. A mechanism has been devised, but not tested, which should give no side forces to the augmented target on release and thus permit accurate target aiming.

For further information, please contact L. Brown (GA).

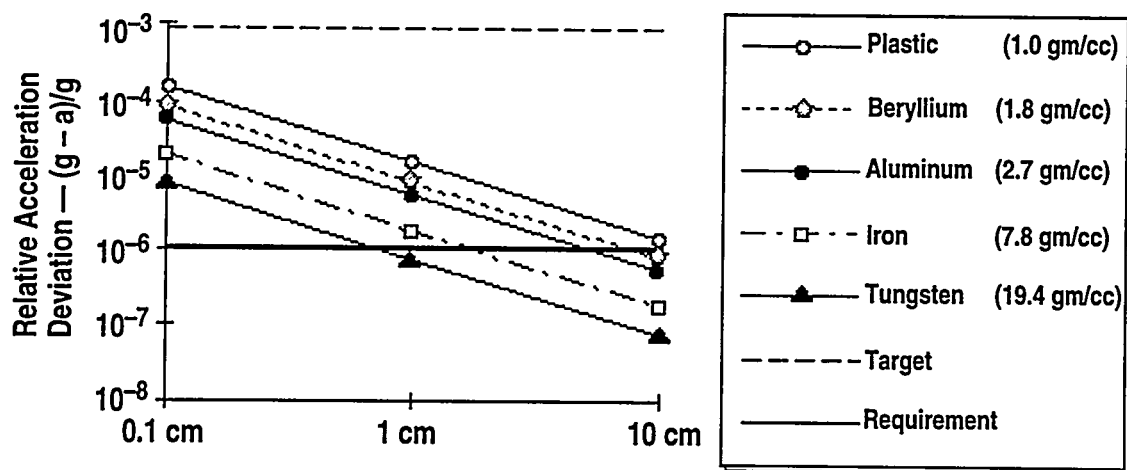


Fig. 6-8. Augmented mass effect on relative acceleration deviation

6.4. REFERENCES

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SECTION 7

PUBLICATIONS

7. PUBLICATIONS

Stephens, R.B. and Wittman, M., "Laser Fusion Target Shell Wall Thickness from Interference Fringe Shape Analysis", *J. Vac. Sci. & Technol.*, (in press).

P. B. Parks and R. L. Fagaly, "Field-assisted Microgravity for ICF Fuel Layering", *J. Appl. Phys.*, (in press).