GA–A23228

CAPSULE PRODUCTION AND DEVELOPMENT FOR ICF EXPERIMENTS

by
A. NIKROO, F.H. ELSNER, D.G. CZECHOWICZ, J. GIBSON, S.E. GRANT,
A.L. GREENWOOD, M.L. HOPPE, D. HUSBAND, B.W. MCQUILLAN,
W.J. MILLER, J.M. PONTELANDOLFO, D.A. STEINMAN,
R.B. STEPHENS, K.R. SCHULTZ, AND M. TAKAGI

OCTOBER 1999

DISCLAIMER

GA–A23228

CAPSULE PRODUCTION AND DEVELOPMENT FOR ICF EXPERIMENTS

by
A. NIKROO, F.H. ELSNER, D.G. CZECHOWICZ, J. GIBSON, S.E. GRANT, A.L. GREENWOOD, M.L. HOPPE, D. HUSBAND, B.W. MCQUILLAN, W.J. MILLER, J.M. PONTELANDOLFO, D.A. STEINMAN, R.B. STEPHENS, K.R. SCHULTZ, AND M. TAKAGI*

This is a preprint of a paper to be presented at the 1st International Conference on Inertial Fusion Sciences and Applications, September 12–17, 1999, Bordeaux, France, and to be printed in the Proceedings.

*Lawrence Livermore National Laboratory, Livermore, California

Work supported by
the U.S. Department of Energy
under Contract No. DE-AC03-95SF20732

GA PROJECT 03748
OCTOBER 1999

ABSTRACT

General Atomics supplies a wide variety of targets for the U.S. Inertial Confinement Fusion (ICF) program. Polymer capsules are mainly fabricated by the depolymerizable mandrel technique. High quality polymer capsules are routinely fabricated in the 400 to 1000 μm diameter range. These capsules may be undoped, doped, or deuterated. This process is under development for fabrication of NIF targets by scaling our current polymer capsule production capabilities up to produce 2 to 3 mm diameter capsules.

1. INTRODUCTION

Our laboratory has been the major manufacturer of U.S. ICF targets and components for a number of years. These include capsules, witness plates, and hohlraums among others. This report is concerned solely with the capsule production and development effort at General Atomics (GA). While a number of different techniques are used for fabricating ICF capsules, the depolymerizable mandrel technique, more often referred to as the glow discharge polymer coated poly-alpha-methylstyrene (PAMS–GDP) technique, has recently come to the forefront of this effort. This method was first used by Letts *et al.* [1] for ICF capsule fabrication. It has been developed since then to fabricate targets on a production scale mainly for ICF experiments at both Nova and OMEGA. We will briefly describe the PAMS–GDP technique in general, followed by more detailed discussions of each of the three steps it involves. We conclude by reporting recent progress in fabrication of National Ignition Facility (NIF) size targets.

2. DEPOLYMERIZABLE MANDREL TECHNIQUE

The depolymerizable mandrel technique involves three steps as shown in Fig. 1. The first step is the fabrication of the spherical mandrel which serves as a mold for the final shell. This mandrel is made of a polymer, poly-alpha-methylstyrene (PAMS), which depolymerizes into gaseous monomers at about 300°C. The PAMS mandrel is then overcoated with glow discharge polymer (GDP) in the second step. In the final step, the GDP-coated PAMS shell is heated to about 300°C under nitrogen. The GDP coating is stable at this temperature while the PAMS decomposes and diffuses out through the GDP. Thus, the remaining capsule is indeed a free-standing GDP shell. This technique allows unparalleled control of wall thickness and composition. Each step of this seemingly simple process has been the subject of extensive development over the past few years. The quality of the final capsule depends heavily on the quality of the product of each step. The starting PAMS mandrel must have the sphericity, concentricity, and surface finish required of ICF targets. The final GDP shell will usually mimic the PAMS mandrel. The GDP coating has to be deposited in a controlled fashion to obtain the desired thickness and surface finish of the final capsule. If dopants are desired in the ablator, they may be added to the GDP film at the desired concentrations.

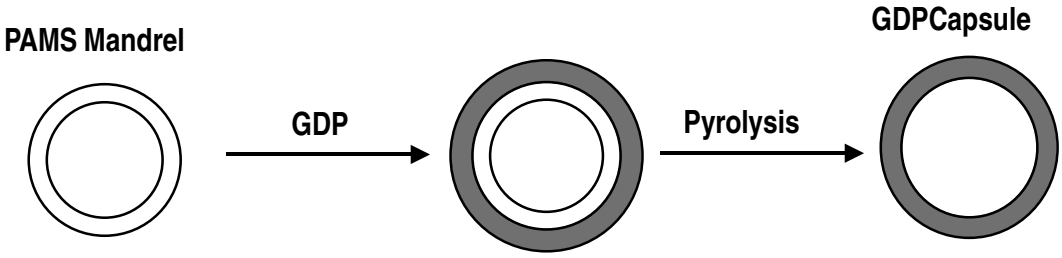

Fig. 1. The PAMS–GDP process involves PAMS mandrel production, overcoating those mandrels with GDP and finally pyrolyzing away the initial PAMS mandrel. The final target is a free-standing GDP capsule.

Finally, the pyrolysis conditions have to be chosen to ensure survival of capsules during this process. The final dimensions of capsules are also influenced by the pyrolysis parameters.

2.1. PAMS PRODUCTION

The idea of utilizing a depolymerizable polymer mandrel for ICF target fabrication was first introduced by Letts *et al.* [1] using PAMS beads. Subsequent research indicated that benefits could be realized by the use of hollow PAMS capsules for GDP coating and pyrolysis rather than beads [2]. These PAMS mandrels are prepared using a method developed at the Institute for Laser Engineering (ILE) in Osaka, Japan [3] which utilizes a triple-orifice droplet generator. An internal aqueous phase, W1, is pumped through the inner needle, and an oil phase, O1 consisting

of PAMS dissolved in an organic solvent, is pumped through the outer coaxial needle. An external aqueous stripping fluid, W2, draws the capsules off of the needle tips as it flows down the collection tube to a collection vessel. Precise control of the W1, O1, and W2 flow rates allows for the production of a large quantity of capsules of uniform diameter and wall thickness. The wet capsules are collected and the solution is heated and agitated until all of the O1 solvent is removed, yielding a water-filled PAMS capsule. The capsules are washed and placed into 20% ethanol to extract some of the interior water phase and to nucleate a bubble in the capsule's interior. The capsules are then dried in a vacuum oven to remove final traces of water before being characterized with regard to sphericity, surface finish, wall thickness, and wall uniformity. Our research efforts [4,5] have found that the physical properties of the final PAMS capsules are sensitive to various factors including: the density mismatch between the W1, O1, and W2 phases; the agitation methods used; the rate at which the capsules are cured; and the additives used in the W2 fluid. Control of these parameters is crucial to the fabrication of high quality mandrels for ICF target fabrication.

2.2. GDP COATING

GDP deposition as applied to ICF target fabrication was initially developed for pure undoped coatings of Nova size targets [6]. In this process, a hydrocarbon gas, referred to as the precursor gas, is broken down in an inductively coupled plasma system. The fragments deposit on the substrates and form a polymer film with a surface finish sufficiently smooth for ICF experiments. The GDP deposition process has been developed substantially over the years to allow the incorporation of dopants in the ablator layer, for coating of larger OMEGA, and more recently NIF size shells. Incorporation of dopants is achieved by using or adding a precursor gas containing the desired dopant atom [2,7,8]. Fully deuterated films may be deposited by simply substituting the process gases by their deuterated versions.

Uniform coating of PAMS mandrels requires agitation during the GDP deposition. A piezo electric shaker has been used with great success for coating Nova size mandrels. The vibrating action of the piezo tube results in the shells skating around in the coating pan. Such an agitation mechanism can also be used for coating larger OMEGA or NIF size shells. However, coating large numbers (10 or more) of such shells, under strong agitation required to obtain uniform coatings, results in frequent collisions between the shells, which leads to coatings that contain large numbers of undesirable domes [9]. To avoid this, two new agitation techniques have been devised. In one method, spinning a slightly tilted pan causes shells to roll, providing enough agitation for uniform GDP coating of mandrels, while avoiding frequent head-on collisions between shells and their detrimental effects. In the third agitation scheme, the coating pan is tapped every few minutes during the coating process. Because of the slow coating rate of the GDP process, this provides sufficient agitation for uniform coating of shells. The mandrels are essentially static for most of the coating, reducing the shell-to-shell collision rate tremendously. Using these techniques, over 30 large (≈2 mm) shells may be coated together with smooth, virtually dome-free GDP films.

2.3. PYROLYSIS

The GDP-coated PAMS shells are pyrolyzed to remove the PAMS mandrel and obtain the final target — a free-standing GDP shell. PAMS depolymerizes at about 300°C. However, it softens and melts around 180 to 220°C. Therefore, in order to minimize the stresses on the GDP layer during pyrolysis and to ensure the survival of the final GDP shell, the heating process must be well controlled between about 200 and 300°C. Shells are held at 300°C for a minimum of 5 h to more than 20 h depending on the thickness of the PAMS mandrel. The entire process may take as long as two to three days. There is considerable shrinkage of the diameter and thickness of the GDP layer during pyrolysis. The diameter may shrink by as much as 6% and the thickness by 10% depending on the length of pyrolysis. This shrinkage is strongly dependent on the type of GDP involved in the process. For example, undoped and deuterated GDP shrink about the same while silicon-doped GDP does not shrink at all [2]. Fortunately, the shrinkage is predictable, and the length of the pyrolysis process may be adjusted to obtain GDP shells within 2% of the desired diameter at the end of pyrolysis.

3. NIF TARGET DEVELOPMENT

Recent progress has been made in scaling up capsule production to fabricate capsules greater than 2 mm in diameter. It is considerably more difficult to fabricate these larger shells with the same sphericity and uniformity of 1 mm shells. However, using a variety of modifications, a number of batches of PAMS mandrels having the desired sphericity and uniformity have recently been made. Figure 2(a) shows a power spectrum, obtained by the AFM spheremapper [10], of the surface of a 2 mm PAMS mandrel from a recent batch. The trace used for many of the theoretical calculations for ICF target performance, dubbed the NIF standard, is also shown for comparison. As can be seen, the PAMS shell has excellent surface mode structure. Many shells in the batch are of the same quality. A number of these shells were used to produce capsules similar to one of the possible target designs proposed for NIF, that is, germanium-doped plastic shells. Figure 2(b) shows the power spectrum of a 120 μm thick GDP shell doped with 3 atomic % germanium. The trace is again very near the NIF standard. These results indicate that future development should produce plastic targets of the desired quality for NIF. GA, in collaboration with Lawrence Livermore National Laboratory personnel, is continuing this work.

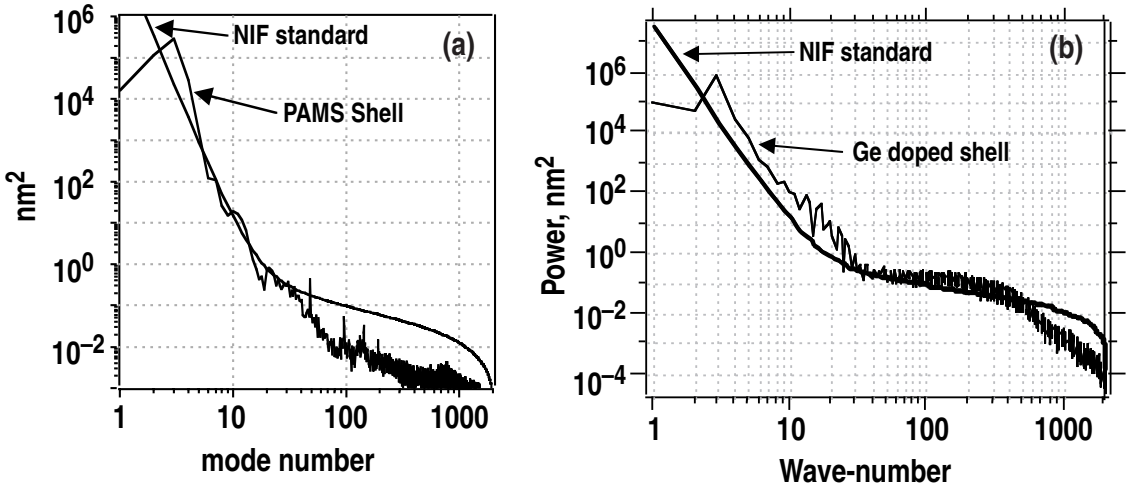

Fig. 2. Near NIF quality GDP capsules have recently been produced. (a) AFM power spectrum of a bare 2 mm PAMS shell, (b) AFM power spectrum of 120 μm thick Ge-doped GDP capsule made using a mandrel similar to one shown in (a). The germanium concentration is 3 atomic %.

4. CONCLUSION

The PAMS–GDP process has been developed into a production process for fabrication of targets for Nova and OMEGA. OMEGA targets often require doped layers at various positions within the shell, including the inner-most layer. The PAMS-GDP process is ideal for the fabrication of such targets. Production and GDP coating of 1 mm shells is now, for the most part, routine. Recent developmental work has concentrated on the fabrication of 2 to 3 mm diameter shells for NIF. Substantial progress has been made in producing polymer capsules which have the desired sphericity, wall uniformity, and surface mode structure desired for NIF targets. Such capsules may be used as starting mandrels for the fabrication of beryllium or polyimide targets as well.

ACKNOWLEDGMENTS

Work supported by the U.S. Department of Energy under Contract No. DE-AC03-95SF20732.

REFERENCES

[1] S.A. Letts, E.M. Fearon, S.R. Buckley, M.D. Sacula, L.M. Allison, R.C. Cook, "Preparation of hollow ICF targets using a depolymerizable mandrel," Mat. Res. Soc. Symp. Proc. **372** (1995) 125.

[2] B.W. McQuillan, A. Nikroo, D.A. Steinman, F.H. Elsner, D.G. Czechowicz, M.L. Hoppe, M. Sixtus, W.J. Miller, "The PAMS/GDP process for production of ICF target mandrels," J. Fusion. Technol. **31** (1995) 381.

[3] T. Norimatsu, C.M. Chen, K. Nakajima, M. Takagi, Y. Izawa, T. Yamanaka, S. Nakai, "Cryogenic targets and related technologies at ILE Osaka University," J. Vac. Sci. Technol. A **12** (1994) 1293.

[4] B.W. McQuillan and A. Greenwood, "Microencapsulation process factors which influence the sphericity of 1 mm o.d. poly (alpha-methylstyrene) shells for ICF," J. Fusion Technol. **35** (1999) 194.

[5] B.W. McQuillan, F.H. Elsner, R.B. Stephens, and L.C. Brown, "The use of $CaCl_2$ and other salts to improve surface finish and eliminate vacuoles in ICF microencapsulated shells," J. Fusion Technol. **35** (1999) 198.

[6] S.A. Letts, D.W. Myers, L.A. Witt, "Ultrasmooth coatings for ICF," J. Vac. Sci. Technol. **19** (1981) 739.

[7] R. Brusasco, M. Saculla, R.C. Cook, "Preparation of germanium doped plasma polymerized coatings as ICF ablators," J. Vac. Sci. Technol. A **13** (1995) 948.

[8] A. Nikroo, "Deposition and characteristics of chlorine doped glow discharge polymer films," J. Fusion Technol. **31** (1997) 431.

[9] A. Nikroo, D. Woodhouse, "Bounce coating induced domes on glow discharge polymer coated shells," J. of Fusion Tech. **35** (1999) 202.

[10] R.L. McEachren, C.E. Moore, R.J. Wallace, "The design, performance, and application of an atomic force microscope-based profilometer," J. Vac. Sci. Technol. A **13** (1995) 983.