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MICROENCAPSULATION STUDIES FOR MASS PRODUCTION OF IFE TARGETS

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We have performed research and development to examine scale-up methods for mass production of IFE targets. We have focused our initial efforts on designing and testing equipment for high yield mass production of plastic capsules. Aspects of microencapsulation identified for improvement include design and fabrication of a stainless steel dual orifice needle to increase reliability of the droplet generator and make easier the process control for future manufacturing methods by providing consistent process flowrates. Additionally, the long cure and dry times (2–4 weeks) required for capsules has been examined, resulting in the initial design of a large, eminently scalable three-phase gas agitated contactor. This apparatus, once functionality has been established, may help begin the transformation of batch scale target fabrication to a continuous process. This research is intended to develop the basis for high-yield, high reproducibility direct drive foam target mass production in future studies.[1,2,3,4]

I. INTRODUCTION

An Inertial Fusion Energy power plant will require an estimated 500,000 targets per day for a 1,000 MW(electric) power plant. A robust, high yield mass production process for the fabrication of IFE targets is needed to reduce manufacturing costs from $2,500 to $0.25 per target. Current laboratory scale batch fabrication techniques, while adequate for research and small quantities (~ 1,000 targets per year) must be reexamined to identify those aspects suitable for mass production.

As an example, poly (α-Methyl Styrene), PαMS, capsules used as mandrels for IFE coatings and experiments are currently produced in a batch process via microencapsulation utilizing a dual orifice droplet generator.[3] As illustrated in Fig. 1, an inner drop of water (aqueous phase) is surrounded by a mixture of PαMS dissolved in fluorobenzene (non aqueous polymer solution).

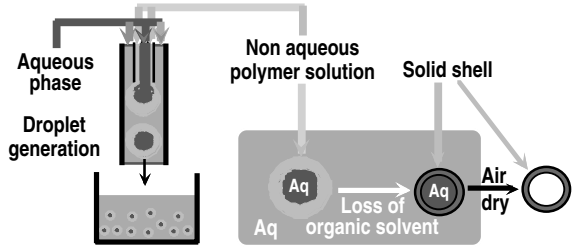

Fig. 1. Schematic of PαMS microencapsulation.

The drop forms by flowing the two solutions through a dual orifice generator, the outlet of which is pictured in Fig. 2. A stripping flow of polyacrylic acid, PAA, in aqueous solution, then strips the drop from the orifice needle at a predetermined rate for a particular size drop (outer aqueous phase). Flowrates vary depending upon the diameter and wall thickness of drop desired, for example, typical flowrates for 2 mm diameter capsules with 20 μm walls are 40 ml/h of inner water and 14 ml/h of the polymer solution, with the outer flow varied to strip the drops at about one per second, usually 3,600 ml/h.

The collected drops, or capsules, are hardened by removal of the fluorobenzene solvent from the non aqueous polymer solution in rotary evaporator contactors warmed in water baths. Once the capsules are solid, they undergo several washes to remove particulate contamination and debris from the capsule surface. At this point, ethanol extraction of the water inside the capsule is performed, followed by approximately two weeks inside a vacuum oven at 25°C to completely dry the capsules.

The dual orifice microencapsulation process is deceptively simple with few process variables to manipulate, however, there is a large influence among these variables when comparing to final capsule characteristics. In the desire to scale up to mass production, the interdependency of the process variables needs to be taken into consideration. For example, if it is desired to

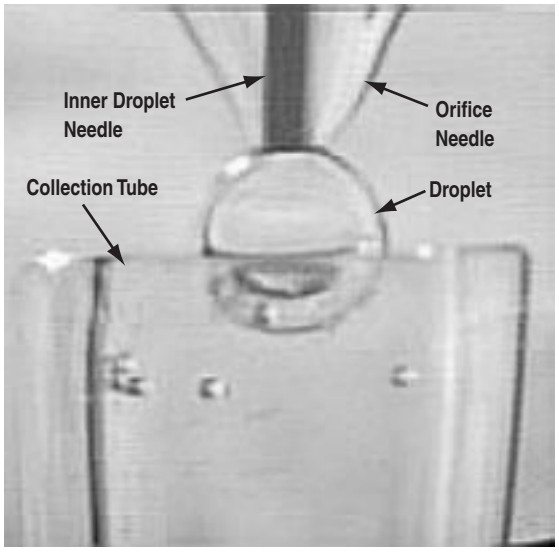

Fig. 2. Droplet formation with dual orifice needle.

shorten the time it takes to remove fluorobenzene solvent from the hardening capsules, thereby decreasing the fabrication time, one could increase the water bath temperature. However, this leads to significant changes in the solubility of the polymer solution in the aqueous phase increasing surface roughness, as explained below, density changes in each phase leading to density mismatch, viscosity changes of the solutions affecting agitation during hardening, and interfacial surface tension changes affecting capsule concentricity. Thus, one process variable change, water bath temperature, can lead to significant alterations in final capsule characteristics.

As mentioned above, the simple desire to increase solvent extraction rates to shorten cure time leads to increased surface roughness. Surface defects are believed controlled by the Marangoni effect Low mode surface roughness has been experimentally correlated[2] with the Marangoni number, M, given by equation:

$$M = \frac{\left(\frac{d\gamma}{dc} \times \Delta C \times L\right)}{\eta \times D} \quad , \qquad (1)$$

In order to reduce these defects, the process variables ΔC, L, and η (concentration gradient, capsule wall thickness, and polymer solution viscosity, respectively) have been manipulated with success in the past to reduce Marangoni convection.[5] However, in the case of larger IFE capsules with 250 μm walls, the Marangoni effect will be amplified by the increased wall thickness, L. Additionally, by attempting to increase the solvent extraction rate, ΔC is adversely increased as well. We must look at not simply ramping up solvent extraction rates, but must look at determining the maximum removal rate that will not destroy the capsules. Experiments are planned to examine methods to alter the variables that control Marangoni cells to reduce surface roughness, including the utilization of new apparatus such as the gas agitated contactor mentioned in Section III, and quantifying maximum solvent removal rates.

II. PROCESS IMPROVEMENT

Mass production of IFE targets differs from the laboratory scale process in significant ways. Our goal is to form at least 500,000 targets per day in a continuous or semi-continuous process.[4] All aspects and assumptions of the batch fabrication process require review to identify those areas that can be altered to increase production. The first such arena to explore is the liquid ratio used in the laboratory scale process. The ratio of total process volume, solution plus wet capsules, to wet capsule volume is 200 for the laboratory process. The ratio will have to be reduced significantly to reach economic target production. One estimate used for calculating quantities of chemicals for large batch runs assumes a ratio of 10 to 1, pending confirmation in experiments that capsules can withstand the reduced liquid ratio. Even with the lower ratio, up to 16,500 L of outer aqueous stripping fluid will be required to produce 500,000 targets using current flowrates. Therefore, we have identified the need for a recycle stream to be implemented in the droplet generation process to reduce costs, as well as further study to determine the minimum liquid ratio the capsules can withstand before damage occurs. The recycle stream will require filtration, and quite possibly regeneration of the flows' chemical composition to achieve consistency in process parameters.

Additionally, parametric study needs to be undertaken to determine the flowrates required for larger IFE targets. While larger diameter targets have been successfully formed in the past (~5 mm diameter), they were formed with a thin wall, ~1% of diameter. Thicker walled capsules as required for IFE direct drive targets (~250 μm, 5.4% of diameter), have not yet been attempted through full cure, but will be tested in the near future. The flow parameters for the thicker walled targets can be estimated with empirically determined correlations of wet capsule diameters with final cured diameters, but will have to be confirmed in the laboratory since a different flow regime than is currently used will be required. Specifically, the ratio of the inner aqueous flow to non aqueous polymer flow is generally 2.8 to 1 for smaller targets (3 mm targets), but may have to altered to at least 1 to 2.3 for the larger 4.6 mm/250 μm wall IFE targets. The effect this will have in the droplet generator and for capsule formation needs be determined, and will require a redesign and modification to the droplet generator apparatus itself for the larger diameter capsules.

III. NEW APPARATUS

Currently, rotary evaporative contactors, Fig. 3, are used to remove solvent from and cure PαMS targets. While these contactors are ideal in curing small batches of capsules, they are difficult to scale-up to larger sizes. The

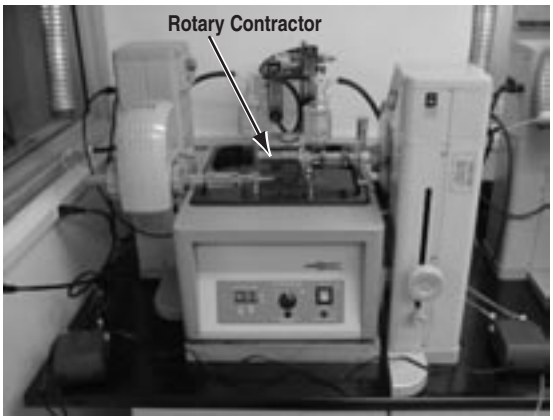

Fig. 3. Rotary contactors for removal of solvent.

change to larger containers has resulted in damaged capsules due to the increased fluid velocities the capsules encounter within the containers. Initial research identified the need for redesign of the contactors to allow for larger batches of capsules to be cured, however, parametric studies have to be conducted with each change in dimension to determine if the new size is feasible and not destroy the capsules. A better design would be one where the apparatus is easily scaleable to larger sizes, with the added benefit of the ability to switch to a continuous process.

We have developed a prototype Gas Agitated Contactor (GAC) pictured in Figs. 4 and 5, to address the need for larger capacity solvent extraction apparatus. The device uses a three phase contact of solvent (fluorobenzene) saturated air, outer aqueous bulk fluid, and semi solid capsules to remove solvent in a controlled, optimized manner. The GAC could possibly replace the rotary evaporative contactor in future mass production scenarios, especially in a continuous process.

The design is eminently scaleable in the X-Y direction, once away from wall effects and holding fluid depth constant, without having to identify new process parameters due to size. An added benefit is easy adaptation of the design to a continuous flow process, ideal for a large

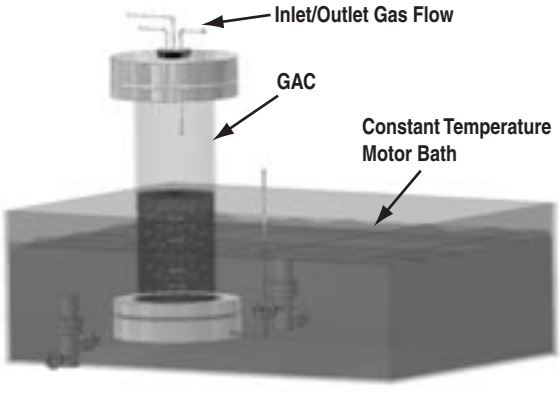

Fig. 4. Schematic of prototype GAC.

Fig. 5. GAC in use. Note the streams of bubbles rising through the bulk fluid. Shells can be seen between the streams.

facility in the future. Functional attributes of the contactor include using bubbles of air to remove fluorobenzene from the system and gently agitate the capsules, as pictured in Fig. 5. Additionally, the inlet gas flow's composition, used to extract the solvent and agitate the capsules, can be controlled via a system of pumps and valves. This allows decoupling of the rate of solvent removal from the capsules with the degree of agitation of the capsules, to optimize curing times. In the event that bubble agitation in the device is at the maximum that can be achieved without capsule damage, yet solvent removal rate (mass transfer rate) is not high enough, a secondary make-up flow can be pumped into the headspace above the bulk fluid liquid level to achieve adequate solvent removal.

This apparatus can also be used for the subsequent washings and water/ethanol extractions without exposing the targets to contamination or the stress of being moved from container to container as is currently done in the laboratory. Initial design and fabrication are complete with test runs under way.

The GAC will require foam minimization in order to be fully functional. We found that while 0.05 wt% PAA aqueous stripping solution, currently used for microencapsulation and curing, minimally foams, 0.3 wt% to 3.0 wt% polyvinyl alcohol, PVA, aqueous solutions used to clean debris off the capsules later in the curing process have significant foaming tendencies. Therefore, a new washing solution with dual roles must be identified that will remove PAA off the capsule wall without foaming and which can be utilized economically in a continuous process. Several solutions have been identified and previously tested for cleaning of the capsules, providing an excellent basis for identifying possible replacement solutions.[3] Takagi noticed that while isopropanol (IPA) did not adequate clean the capsules; neither did it appear to damage the capsules once the capsules were semi-cured. We set up an experiment to intentionally foam a 3.0 wt% PVA aqueous solution with a bubble stream. As the foam layer began to form, small IPA drops were sprayed onto

the foam, resulting in foam dissipation. This method will be tested in conjunction with the GAC in future experiments to confirm results and ensure the capsules are not affected by the addition of minimal IPA to the bulk aqueous solution used for curing.

Another device identified for modification is the droplet generator apparatus currently utilized in the microencapsulation process. A handmade glass needle is currently employed in the system, previously pictured in Fig. 2. Several shapes, with convex to concave outer cross sections, of this needle are in use with each one requiring slightly different flowrates to achieve a capsule with similar characteristics and dimensions. This is not conducive to mass production, as it would be expensive and time-consuming to identify and reestablish flowrates every time a fragile glass needle required replacement. Therefore, we have replaced this particular element of the droplet generator with a stainless steel needle, pictured in Figs. 6 and 7.

The needle is stainless steel as well as self-centering to provide for easy, consistent fabrication and replacement. It is anticipated that designs with slight variations will need to be tested to ensure adequate operation, but once defined, will provide an improvement in repeatability and consistency in the use of droplet generators in mass production of IFE targets. Initial tests with the apparatus pictured in Figs. 6 and 7 resulted in slightly smaller PαMS targets (~74%–90%) in diameter than with glass needle designs, therefore, flowrates are being established for the new design.

IV. FUTURE WORK

Near term mass production goals for microencapsulation research and development include the following: identify and confirm flowrates for larger diameter and thicker walled targets, examine alternative washing solutions or processes for use in the GAC, and optimization of solvent removal rates within the new GAC apparatus.

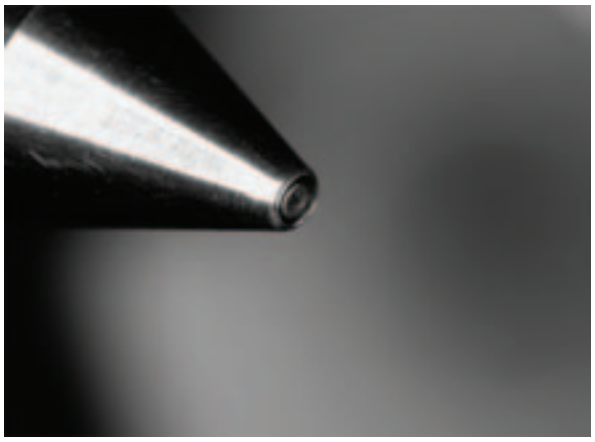

Fig. 6. Self-centering metal needle for droplet generator.

Additionally, longer term research will focus on adding process control to microencapsulation to increase the yield of acceptable targets. This will most likely entail real-time optical imaging and analysis systems to characterize target diameter and wall thickness of wet capsules (once robust correlations between wet capsules and cured capsules have been established). Other process improvements include adding a pulse system to the droplet generator to better control capsule formation, increasing production rates and possibly reducing the amount of debris collected in the bulk fluid. Lastly, forced convection is being considered to decrease the long drying times associated with curing the capsules, a prime opportunity to reduce target production time.

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Fig. 7. Inner self-centering needle showing the square centering fins (~0.05 inch diameter).