

DEVELOPMENT OF WATER CAPSULES

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By

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FOREWORD

This report was prepared by Southwest Research Institute, San Antonio, Texas, under USBM Contract Number HO272042. The contract was initiated under the Coal Mine Health and Safety Program. It was administered under the technical direction of the Spokane Mining Research Center with Robert E. Simpson acting as Technical Project Officer. Larry Rock was the contract administrator for the Bureau of Mines. This report is a summary of the work recently completed as a part of this contract during the period September 1977 to October 1978. This report was submitted by the authors in October 1978.

ABSTRACT

This project had as its objective the experimental investigation of water capsule production utilizing the Southwest Research Institute's submerged nozzle encapsulation apparatus (U.S. Patent 3,389,194). Initial efforts on this project involved nozzle configuration and shell formulation optimization. The shell formulation developed on a prior program was modified during the course of the project to obtain the best balance of capsule breakage resistance and water retention.

Subsequent effort centered on determining operating conditions which would produce the best quality capsules for each candidate shell formulation as determined by laboratory tests. Some preliminary work was done to increase the production capacity of the encapsulation equipment. The program was extended with major emphasis concerned with scale-up studies of capsule production capacity and the treatment of problems associated with grouting cartridge fabrication.

During the course of the program, water-containing capsules were produced which have good long-term water retention, block resistance and adequate strength. Using a single-nozzle design, a production capacity of 15.9kg (35 pounds) per hour was demonstrated to give acceptable capsules based on initial results.

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I. INTRODUCTION

In a concept originated by the U.S. Bureau of Mines' Spokane Mining Research Center for a new inorganic-grout mine-bolt system, it was envisioned incorporating water-containing capsules in a fast-setting cement such that the capsules in the mix could be ruptured at the proper time by mechanical pressure and shear of the mix, and the released water would set the cement. Under a feasibility program, water-containing capsules prepared for the Bureau of Mines by SwRI demonstrated considerable promise. In order to further evaluate the concept, SwRI was contracted by the Bureau of Mines to investigate the preparation of water-containing capsules which could be studied in the development of a practical system.

II. OBJECTIVE

The objective of this program was the experimental preparation of water capsules utilizing the Southwest Research Institute's submerged nozzle encapsulation apparatus and the evaluation of certain characteristics of the capsules which would be of significance in the use of the capsules for a fast-setting cement.

III. EXPERIMENTAL

A. Encapsulation Apparatus

Southwest Research Institute's submerged nozzle encapsulation apparatus, the basic design of which is illustrated in Figure 1, consists essentially of a nozzle of two concentric tubes mounted axially in a tubular duct. Capsule filler material is pumped through the center tube and fluid shell formulation through the annulus of the nozzle so as to extrude a fluid "rod" of filler encased in a sheath of the shell formulation in a carrier fluid. Subsequently, the extruded rod is broken into individual capsules. The capsules formed by the break-up of the extruded rod are transported down the carrier fluid duct for subsequent hardening of the capsule shell, and the capsules collected on a screen prior to recirculation of the carrier fluid.

Feasibility capsules prepared prior to initiation of this project were prepared on a laboratory device that employed a coiled-jacketed cooling section in the duct into which the nozzle discharged the shell and fill streams for capsule formation. These nozzles were determined not to be correctly sized nor was the device suited to high production rates of capsules of the size required for the intended application. The device also contained a sight glass for observation of capsule formation that introduced some turbulence in the zone of capsule formation. A straight-run jacketed duct was therefore fabricated for this project to eliminate problems with the coiled configuration and the sight glass section was replaced with a stainless steel convergence section that provided a smooth transition of fluids to the jacketed duct.

B. Formulation and Evaluation Studies

1. "Basic" Shell Formulation

The examination of candidate capsule shell formulations was conducted concurrently with changes in the encapsulation system to improve system performance and increase production rate; however, the capsule shell formulation and capsules made with the various formulations were considered independently of the conditions under which the capsules were formed.

Initial encapsulation runs were made using deionized water as capsule fill and tap water as carrier fluid with the "basic" shell formulation that was used to prepare the initial feasibility capsules. This "basic" shell formulation had been developed on a prior program for water encapsulation. The formulation is:

47 percent Piccolyte S-115 (Hercules, Inc.)
40 percent Sunoco 4412 wax (Sun Oil Co.)
13 percent Polybutene 128 (Chevron Chemical Company)

A major deficiency of the formulation was the low temperature at which capsules made with it stuck together (blocked). With insufficient cooling, blocking also occurred during collection of the capsules. In addition, the tackiness of the formulation in the molten state tended to cause coating of the carrier tube of the encapsulation device and resulted in plugging that prevented extended runs. The strength of these capsules was low, and therefore an increase in resistance to rupture was considered desirable.

During the program, the "basic" formulation was used to prepare capsules under a variety of conditions. Twenty-three of the encapsulation runs are described in Table I (these runs are indicated by one asterisk in the table) which gives data for all significant encapsulation runs made during the program. Analytical data for the runs are included in Table II (these runs are also indicated by one asterisk).

Input pump rates for the "basic" formulation runs were set to give payloads of 63.8 percent to 65.8 percent in 21 of the 23 runs listed in the table, and capsule analysis gave payloads of 60.2 to 64.8 percent for capsules from the runs. In the other two runs (2-22C and 2-24A), input was set to give capsules of 69.2 percent payload and analysis showed 69.3 and 68.0 percent.

Capsules from 16 of the runs were tested for weight loss by exposing samples to the atmosphere at ambient temperature (67 to 75°F) and relative humidity (45 to 60 percent). Results are presented in Table II. Losses are given as percentage of the original fill lost during the first three days, during the first seven days, and the average per day for an additional number of days as indicated. The capsules were dried overnight by

INTERNAL
PHASE

SHELL
FORMULATION

CARRIER FLUID

FIGURE 1.

SUBMERGED EXTRUSION NOZZLE



exposure to the atmosphere before samples were taken to analyze and test. Surface moisture and water fill from capsules with cracks or holes should be lost during the first seven days, and the loss thereafter should give an indication of water loss through the shell. The average weight loss of the original capsule fill after the first seven days for 13 of the 16 runs with the basic shell (omitting 1-23A, 1-23B and 1-38A as not being representative because capsules were damaged by blocking on collection) was 0.019 percent per day for capsules on test from 45 to 148 days. The range was 0.091 to 0.001 percent per day, and average deviation from the mean was 0.020 percent. The average total loss for the first seven days was 3.7 percent.

The rate of loss for an individual sample of capsules generally decreases with time and rate of loss would be expected to increase with payload, but these effects are not significant at the low loss rates of these capsules. A trend toward higher loss rate with decrease in capsule size is indicated. In this connection, it should be noted that the capsules are not all single centered spheres, but a varying percentage of the larger capsules are multicore, with two, three or more droplets of fill. Most of the capsules prepared using the basic formulation were made before equipment was available to allow formation and collection of small capsules made at higher production rates.

Break strength of samples from eight runs tested averaged 24 grams for single capsules in the 1700- to 1410- micron size range. In the test, about 20 single capsules are crushed against a rigid surface with a mechanical force gauge, and the results averaged. The simple test is of use in preliminarily comparing capsules made with different shells. The validity of the test as a gauge of bulk capsule resistance to damage in handling was not determined.

Capsules from Run 2-22C, made with the "basic" shell formulation and having a payload of 69.3 percent, were tested for break-strength relationship to size with the following results:

| Size (microns) | Wt. % of Sample | Break Strength (grams) |
|----------------|-----------------|------------------------|
| 2800-2360 | 13.2 | 47 |
| 2360-2000 | 33.0 | 21 |
| 2000-1700 | 35.2 | 11 |
| 1700-1410 | 10.1 | 16 |
| 1410-1180 | 4.4 | 15 |

Aluminum stearate was added to the basic shell formulation in one encapsulation run (Run 1-16) and a fatty amide—Armid HT—to the formulation in four runs (1-7, 1-28, 1-40 and 1-43C). The stearate had little apparent effect, but the Armid HT inhibited blocking to a small extent and aided in the formation of smaller capsules to a minor degree.

2. Experimental Formulations

a. Formulations Containing Elvax

The first four formulations (1-9 through 11-3, Table I), tested as possible substitutes for the "basic" formulation, contained ethylene vinyl acetate copolymer (Elvax 210) in mixtures with the paraffin wax and Piccolyte resin used in the basic formulation. The material added strength to the capsules and reduced blocking but severely increased loss in both initial and long-term tests (Table II). Average seven-day loss for the capsules from the runs was 12.4 percent of the original fill. The average loss rate after the first seven days was 0.341 percent per day (64 to 87 days). The average break strength was 60 grams.

b. Formulations Containing Ethylene-acrylic Copolymers

Two ethylene-acrylic copolymers were used with paraffin wax to make four encapsulation runs (1-12B, 1-12C, 1-15A and 1-15B, Table I). The ethylene-acrylic copolymers composed only five percent of the formulations because of the high viscosity of the materials at use temperature. The strength of capsules from the three runs tested was greater than that of the basic capsules, but loss of fill was high, although better than the capsules made with Elvax 210 (Table II). Break strength averaged 37 grams. The first seven-day loss

of fill averaged 21.9 percent for the three runs, and the average loss rate after this was 0.236 percent per day (80 to 84 days).

Two runs (1-18A1 and 1-18A2) were made with a shell formulation of paraffin, Piccolyte and ethylene-acrylic copolymer (Gulf 2205). Break strength for the sample of capsules tested from Run 1-18A2 was 32 grams, but loss rate was high (0.199 percent per day for 73 days after the initial seven-day loss).

c. *Formulations Containing Polyethylene*

Polyethylene, as a component, was tested extensively in mixtures of polyethylene-paraffin, polyethylene-paraffin-Piccolyte, and polyethylene-paraffin-Piccolyte-polybutene. The runs were made at low production rates because the increased temperature requirements could not be met at high rates by the heating system available at the time.

(1) *With Paraffin Wax*

Only one run (1-14, Table I) was made with a formulation containing polyethylene and paraffin wax. The low molecular weight polyethylene, Epolene C-16, was used in the formulation with Sunoco 4412. Capsules from the run had a break strength of 67 grams, but although the capsules looked good, initial loss was very high, and long-term loss was high (0.220 percent per day average after the first seven days for 81 days, Table II).

(2) *With Paraffin-Piccolyte*

Fifteen encapsulation runs were made using polyethylene with paraffin wax and Piccolyte S-115 resin in the shell formulations, with fill loss rate average of 0.060 percent per day for 152 to 153 days from capsules in two runs (2-12B and 2-14A) approaching that of the basic formulation (runs are indicated by two asterisks in Tables I and II). The shell formulation used for Run 2-12B was made with a higher melting paraffin wax (Boler 1426) and only five percent low molecular weight polyethylene (Epolene C-16), but capsule break strength was 62 grams and the capsules did not block. The formulation for Run 2-14A was the same as 2-12B but contained a mixture of lower melting paraffin (Sunoco 4412) with a higher melting paraffin (Boler 1426), and this resulted in capsules with break strength no higher than that of the "basic" formulation capsules. The other formulations made with polyethylene, paraffin and Piccolyte were similar to the "basic" formulation but with Epolene C-16 low molecular weight polyethylene substituted for polybutene and some change in the ratio of components. The paraffin used was Sunoco 4412, and the Piccolyte was S-115. Capsules from the twelve runs tested gave an average break strength of 46 grams, but loss rate was high, averaging 0.188 percent per day after the first seven days for 56 to 73 days.

(3) *With Paraffin-Piccolyte-Polybutene*

Capsules from several encapsulation runs made with shell formulations containing polyethylene with paraffin, Piccolyte and polybutene gave long-term loss rates as good as capsules made with the "basic" formulation but with higher break strength and increased resistance to blocking. Four of the samples of capsules tested (from Runs 1-51C2, 1-56C, 2-2 and 2-14B, Tables I and II) gave an average loss per day of 0.025 percent after the first seven days for 123 to 152 days. Because of heating and cooling deficiencies in the system at the time the capsules were made, capsules were not optimum, and this was reflected in high loss of fill for the first seven days for samples from Runs 1-51C2 (10.9 percent) and 2-14B (9.3 percent). Average loss rate for the other five samples tested (from Runs 1-20B1, 1-27B1, 1-27B2, 1-35A1 and 1-46) was 0.149 percent per day after the first seven days for 51 to 153 days. All of the formulations but two contained Sunoco 4412, Piccolyte S-115, Polybutene 128, and Epolene C-16. Run 2-2 contained Sunoco 5512 as well as Sunoco 4412, and 2-14B contained Boler 1426 with Sunoco 4412 as the paraffin wax component.

One additional shell formulation (Run 2-26) contained two percent carnauba wax and three percent C-16 polyethylene with Sunoco 5512 paraffin, Piccolyte and polybutene. Capsules from this run had good block resistance and low fill loss rate (1.0 percent for the first seven days and 0.018 percent average per day for 125 days).

d. *Formulations Containing Higher Melting Paraffin Wax*

Two shell formulations containing a very high melting paraffin wax, Shellwax 700, were used to make five encapsulation runs. The runs were 1-32B1, 1-32B2, 1-55A, 1-55B and 1-55C as listed in Tables I and II. Twenty percent Shellwax 700 was used with Sunoco 4412 wax and Piccolyte in Runs 1-32B1 and 1-32B2. Average seven-day loss was 23.2 percent, and average loss per day was 0.28 percent after the first seven days. Capsules from the other runs tested were much better, but the formulation contained only five percent of the Shellwax, and polybutene was included. Capsules from Run 1-55B gave a loss rate of 0.013 percent per day for 127 days after the initial seven-day loss and had a break strength of 32 grams.

e. *Formulations Containing Wax Substitution*

Samples of capsules from seven encapsulation runs were tested in which Sunoco 5512 wax was replaced in the final selected formulation, as described in Section III.B.3.a, with paraffin waxes of comparable grade (Boler 1435 and Boler 1426). Fill loss rates for capsules from these runs were not very different than those made with Sunoco 5512 wax. Average percent loss per day was 0.016 for 131 to 158 days. First three-day loss per day was 0.9 percent, and first seven-day loss was 1.2 percent. Break strength averaged 21 grams. The capsules blocked more readily than did the Sunoco 5512 capsules, as the Boler wax was tackier. In one run (2-21A) with Boler wax, polybutene was eliminated, resulting in capsules that resisted blocking better but which gave higher loss rate —0.040 percent per day for 153 days.

Two samples of capsules (2-20A and 2-20B) made with the Boler wax substituted for Sunoco 5512 showed small increases in weight after being on test for an extended period. Tests made showed that if oxidation was occurring it was not enough to cause significant masking of loss of fill, and the small difference was within experimental error.

3. *Formulation of Choice*

a. *Composition and Experimental Conditions*

The capsule shell formulation selected as being most practical and best meeting requirements for intended use is as follows:

| Materials | Percent by Weight |
|--------------------------|-------------------|
| Sunoco 5512 paraffin wax | 45 |
| Piccolyte S-115 | 45 |
| Polybutene 128 | 10 |

Experimental conditions were the following:

| | |
|--------------------|---|
| Nozzle: | Outer orifice 0.155 inch |
| Fill tube: | 0.095 inches OD x 0.077 inches ID |
| Carrier flow: | 10 liters per minute at 78°C with temperature at recovery about 38°C |
| Carrier fluid: | Tap water with 0.05 percent Polyox WSR-301 |
| Shell temperature: | 96°C |
| Fill: | Deionized water with 0.05 percent Polyox WSR-205 plus dye (dye aids visual monitoring of quality) |
| Fill temperature: | 73°C |
| Fill rate: | 205 grams per minute |
| Shell rate: | 106 grams per minute |
| Capsule payload: | ca. 65 percent |

During the program, over fifty encapsulation runs were made with the shell formulation of choice, and samples of capsules from most runs were tested for weight loss and size-distribution. Few of the runs were made under the same conditions since the runs were made concurrently with equipment development and scale-up. Payload, size range, and duration of tests for fill loss varied; however, a good estimation of capsule fill retention is shown by the water loss data collected. Antioxidant, butylated hydroxytoluene, was added (0.02 percent) to the shell in four runs (2-36B, 2-37A, 2-37B and 2-38), but no effect on the capsules could be noted.

The conditions of preparation and the results of evaluation of the capsules for certain of the runs are presented in Tables I and II. (The runs are indicated by a triple asterisk.)

b. *Weight Loss*

Samples of capsules from the first 30 encapsulation runs made with the formulation were tested for weight loss for 31 to 181 days after the initial seven days. Original fill (payload) of the capsules from the 30 runs averaged 65.7 percent with a range of 59.4 to 71.2 percent. Average loss of fill per day as percent of the original fill for capsules from the 30 runs was 0.017 percent, with a range of 0.006 to 0.041 percent per day and an average deviation from the mean of 0.007 percent. Average total loss of fill for the first three days on test for capsules from the 30 runs was 1.1 percent of the original fill, and for the first seven days, loss was 1.3 percent. As with other capsules, samples were drained and dried by exposure to the atmosphere overnight prior to sampling.

c. *Capsule Size Distribution*

Capsule size distribution varied, with from 10 to 83 percent by weight being larger than 2,000 microns. Neither the payload nor size had an obvious consistent effect on the capsule water-loss rates.

d. *Break Strength*

Break strength of the capsules made with shell of the selected formulation averaged the same as that of capsules made with the "basic" formulation—24 grams for capsules of about 65 percent payload. Capsules with higher payload gave lower break strength. Since no problem in handling and processing the capsules due to inadequate strength occurred, it was concluded that the strength was adequate for the intended use.

4. *General Evaluation*

a. *Capsule Size*

While capsules submitted for use have apparently been within a usable range, capsule size limitations must be established by use requirements. Capsule size distribution has been reported in Table II as weight percentage above 2,000 microns, percentage from 1,000 to 2,000 microns (or 1,180 to 2,000) and percentage under 1,000 microns (or 1,180). Capsules were sieved into eight fractions for most runs, including three fractions above 2,000 microns. In runs where sizable fractions were over 2,000 microns, most capsules were in the 2,000 to 2,300 micron range, a few percent by weight were in the 2,300 to 2,800 micron range, with no more than about one percent over 2,800 microns. The larger capsules were almost always multicore. At high production rate, shifting capsule size toward a small diameter appeared to result in an increase in satellites.

b. *Blocking*

Tests were made to determine resistance to blocking of capsules made with several shell formulations. Samples on test for almost five months were examined. The capsules were held in PVC plastic pipe of two-inch inside diameter to a height of 34 inches to simulate storage in a 55-gallon drum. No water was added to the capsules which were initially free-flowing. About a kilogram of capsules per test was poured

into each pipe, and the ends of the pipes were closed with rubber stoppers. The pipes were held vertical at room temperature (ca. 75°F) during the course of the test. Samples tested were as follows:

1. Capsules from Run 2-8, made with the selected formulation.
2. Capsules from Run 2-21, made with Boler wax 1426 substituted for Sunoco 5512 in the selected formulation.
3. A mixture of capsules from Runs 2-22C and 2-24A made with the basic shell formulation.
4. Capsules from Run 2-26 made with the formulation containing carnauba wax and polyethylene

Capsules prepared from all but the "basic" shell formulation were freed easily by tapping the pipes and were free-flowing, although damp, with no lumps or apparent damage. The samples of capsules having the "basic" shell were more difficult to free, and large lumps of capsules came out of the pipe with free-flowing capsules. The lumps could be parted easily with no apparent damage, however.

Data for additional blocking tests and details of the test are included in Appendix B. Tests showed good blocking resistance for capsules made with the selected formulation (Run 2-8) with little or no blocking occurring with exposure of 30 minutes at temperatures under 48°C (118°F). Blocking occurred at 48-53°C (118-127°F), but capsules parted with little or no damage when cooled; however, blocking and damage occurred at 56°C (133 F) and above. Blocking resistance was lower when Boler 1426 paraffin wax was substituted for the Sunoco 5512. Some capsules containing polyethylene, such as those from Run 2-12B, gave superior blocking resistance. Capsules made with the basic formulation (1-53A) blocked at 43-46°C (109-115°F) but could be parted. Obvious damage to these capsules occurred at 47°C (116.6°F).

Several agents that might be used to inhibit blocking were tested at 54°C, but only one of the materials (micronized silica) showed any beneficial effect.

C. Scale-up and Process Optimization Studies

1. Turbulence Reduction Aids

Initial encapsulation runs on the project were made at a production rate of 3.26 kilograms (7.2 pounds) of capsules per hour using the "basic" formulation; however, the material plugged the encapsulation system in extended runs. Samples were not recovered, and runs are not listed in tables. Elvax shell formulations were used at a rate of production of 2.22 kilograms per hour for extended runs with no plugging; however, capsules were poorly formed. When Polyox WSR-35 polyoxyethylene was added to the carrier water as a turbulence-reducing agent, capsules were much improved. The Polyox WSR-35 and a more effective successor, Polyox WSR-301, were therefore used throughout the project. No further problem with plugging was encountered in the carrier fluid during the course of the project, even with the basic formulation.

2. Nozzle Design

Pending fabrication of a new nozzle, the initial runs (Runs 1-9 through 1-27A in Table I) were made with an old nozzle drilled out to conform to specifications designed to make shell, fill and carrier flow more compatible at higher production rates. The original nozzle shell orifice was 0.155 inches ID and the fill tube was 0.031 ID × 0.050 inches OD. The new nozzle was 0.155 inches ID with a fill tube 0.028 inches ID × 0.042 inches OD. Many candidate shell formulations were tested with the nozzles at production rates of 2.21 kilograms per hour to 3.29 kilograms with carrier rates of 2.7 to 4.5 liters per minute at 64 to 83°C with capsule payload about 65 percent. No trouble was encountered obtaining cooling and heating necessary for the shell formulations, and well-formed capsules were made with only a small weight percentage being over 2,000 microns in size.

The nozzle was further changed by increasing the fill tube to 0.077 inches ID × 0.095 inches OD with the outside orifice remaining at 0.155 inches. This was the final nozzle configuration and was used beginning with Run 2-37A.

3. Orifice Design

The fill tube orifice was positioned at about 1/8 inch to about 1/32 inch in back of the nozzle orifice with the best setting apparently about 1/16 inch in back of the nozzle orifice. The variations are noted under "Comments" in Table I.

4. Fill Tube Design - Carrier and Cooling Water Variations

Fill tube size was increased to 0.064 inches OD \times 0.043 inches ID beginning with Run 1-32A1, resulting in some improvement in capsules in that fewer multicore capsules were formed. When production rate of capsules with the "basic" shell formulation was increased to 9.1 kilograms per hour with carrier flow rate at 4.4 liters per minute, a high percentage of capsules were larger than 2,000 microns, and the cooling system was insufficient to prevent blocking or sticking of the capsules on collection. Cooling water was supplied to the system at pressure and temperature (76°F) of the laboratory tap water supply pending delivery of a chilling system. A surge tank was added to the recovery system, and this allowed an increase in carrier flow to about 5.58 liters per minute with a production rate of 9.08 kilograms per hour. Some reduction in capsule size was noted but without severe sticking of the capsules.

For additional studies, the inner fill tube was changed to 0.081 inches OD \times 0.062 inches ID to further improve capsules. When carrier flow was increased to about 6 liters per minute at a temperature of about 60°C, surge tank temperature was about 40 to 41°C which caused capsules made with the "basic" shell formulation to stick together badly. At a carrier flow of 7 liters per minute at a temperature of 64°C and a collection surge tank temperature of 42°C, capsules formed well and were recovered without sticking using the selected shell formulation at a production rate of 9.08 kilograms per hour. For a sustained run, collection surge tank temperature increased to 46°C, and slight sticking did occur. Severe sticking occurred with Boler 1426 substituted for Sunoco 5512 in the formulation.

Beginning with Run 2-20A, a booster pump was introduced into the cooling water line to increase flow through the cooling jacket. This allowed cooling of the carrier fluid to 39°C as measured at the collection point with a flow rate of 8 liters per minute at 65°C. Capsules were recovered without sticking under these conditions at a production rate of 10.22 kilograms per hour with the selected shell as well as with the Boler 1426 wax substituted in the formulation. The heating system could not maintain a carrier temperature of 68°C under these conditions, and higher melting formulations therefore could not be used.

With Run 2-22C, a chiller was installed in the system and the collection tank temperature was maintained at 30-32°C for Run 2-22C with carrier flow rate of 7.5 liters per minute at 60°C. Capsules were made with the "basic" shell formulation at the rate of 10.22 kilograms per hour with no sticking or problems during the run. This was the maximum rate tested for the formulation. Carrier temperature could not be maintained above 62°C with the increased cooling under the conditions used.

Beginning with Run 2-35, the encapsulation equipment was moved into a new building and redesigned with an adequate steam supply. An additional heat exchanger was introduced into the carrier flow system so a high carrier temperature could be maintained with high flow rates.

With adequate heating and cooling available, input production rate using the selected shell was increased with runs made at up to 27.2 kilograms per hour. Carrier flow was as high as 11.3 liters per minute and carrier temperature as high as 83°C.

5. Material Balance

Rate of capsule recovery at higher production rates did not equal the production rates estimated by fill pump and shell pump calibration. This was attributed to pump slippage, imperfect capsule formation resulting in lower payload than estimated, and the increased amount of satellites formed. (Satellites are small particles of solid shell which form as the extruded rods of shell and fill form the capsules.) A filter was placed in the carrier stream after capsule recovery to remove the satellites and prevent their recirculation and remelting in the heat exchanger where they would collect and eventually disrupt operation. In two runs (Run 3-1B and 3-4B), satellite recovery indicated that loss of capsule production due to satellite formation was about 3.5 percent and 4.9 percent.

6. *Capsule Quality vs. Production Rate*

Capsule quality appeared to diminish as production rate exceeded a recovery rate of about 15.9 kilograms (35 pounds) per hour. Above this rate, capsule payload was less than theoretical based on pump rate settings, and initial weight loss rates of these capsules indicate poorly formed capsules. No difficulty was encountered in maintaining production at 22.7 kilograms (50 pounds) per hour; therefore, an extended effort was made at this rate, without success, to improve capsule quality to an acceptable level. Since capsules at this rate are marginal, some change in shell or some equipment refinement may overcome the problems.

IV. SUBJECT INVENTIONS

No invention disclosures or patent application have been made by Southwest Research Institute as the result of work done under this contract.

V. CONCLUSIONS

- A. Water-containing capsules can be prepared using SwRI's submerged nozzle process which have good long-term water retention, block resistance and adequate strength.
- B. The capsule shell formulation which gave the best overall characteristics consisted of the following:
- 45% Sunoco 5512 Paraffin Wax
 - 45% Piccolyte S-115
 - 10% Polybutene 128
- C. Using the above formulation, acceptable capsules can be produced at a rate of 15.9kg (35 pounds) per hour using a single-nozzle system. Approximately 148 pounds of capsules were produced using the formulation and forwarded to the Spokane Mining Research Center.
- D. The submerged nozzle encapsulation apparatus utilized on this program is readily amenable to scale-up to commercial production. A single-nozzle unit was operated in the development program, and scale-up is merely a matter of providing a multiplicity of parallel nozzles to achieve the desired production capacity. Common fill and shell tankage, carrier fluid heating and cooling facilities, and finished product handling equipment would be used for all nozzles. Regarding the economics of capsule production, capsule unit costs are obviously lower with increased production rates. The labor cost, which can be considered as a fixed cost per operating hour for a given size installation, is generally the dominant factor in the unit cost at low production levels and decreases in importance as the production rate rises. Shell material and utilities costs are essentially proportional to production rate and are therefore a constant in the unit cost and become the dominant factor at high production rates. Depending upon the possibility of utilizing operating labor on other tasks within the manufacturing organization, it would be economically attractive to operate the encapsulation equipment at high rates on an intermittent basis to minimize the labor cost factor. This alternative should be considered in arriving at optimum plant size.

APPENDIX A
Tables

Table I
ENCAPSULATION RUNS

| Run No. (SwR: No.) ¹ | Shell Composition | % | Estimated Payload wt. % | Estimated Production kg/hr | Carrier Rate mL/min | Carrier Temp. °C | Comment |
|------------------------------------|--|----------------|-------------------------------|----------------------------------|---------------------------|------------------------|---|
| 1-9 (7-032) | Sunoco 4412 wax Piccolyte S-115 Elvax 210 | 60 25 15 | 64.9 | 2.22 | 2800 | 65-69 | Ran about 14 hours without plugging. Capsules did not stick together. Capsules were not well formed. Some bad capsules were removed by floatation after standing overnight. 36 pounds were shipped. |
| 1-10A | Same as 1-9 | | 64.9 | 2.22 | 4000 | 65-69 | Capsules looked better during production than 1-9, but not well formed. |
| 1-109 | Same as 1-9 | | 64.8 | 3.29 | 4500 | 65-68 | Capsules poorly formed. Do not look as good as 1-10A. |
| 1-11E (7-033) (7-034) | Same as 1-9 | | 64.8 | 3.29 | 4500 | 65-68 | 0.5% Polyox WSR-35 was added to the carrier water to reduce turbulence. Capsules appeared much better on production with almost no empty capsules apparent. |
| 1-12B | Shellwax 200 Gulf P.E. 2205 | 95 5 | 66.9 | 2.36 | 3000 | 78-80 | Very few empty capsules when produced. Appeared better than preceding four runs. 0.5% Polyox in carrier. |
| 1-12C | Same as 1-12B | | 66.9 | 2.36 | 2700 | 78-83 | Not quite as well formed as 1-12B. Very few empties when produced. Polyox used. |
| 1-14 | Sunoco 4412 wax Epolene C-16 | 80 20 | 64.8 | 2.21 | 3000 | 80 | Formed well. No empties when produced. Looked good. Polyox used. |
| 1-15A | Sunoco 4412 wax Union Carbide DPDA-2304 | 95 5 | 64.8 | 2.21 | 3000 | 81 | Formed well. No empties when produced. Looked good. Polyox used. |
| 1-15B | Same as 1-15A | | 64.8 | 2.21 | 2700 | 81 | About the same as 1-15A. Polyox used. |
| 1-16 | Sunoco 4412 wax Piccolyte S-115 Polybutene 128 Aluminum stearate 0.5% of above added | 40 47 13 | 64.8 | 2.21 | 3000 | 64-65 | Made well with only a slight tendency of capsules to adhere together when warm. No empties when produced. Some multicores. Polyox used. |
| 1-17 | Sunoco 4412 wax Piccolyte S-115 Polybutene 128 Armid HT - trace | 40 47 13 | 64.8 | 2.21 | 3000 | 64-65 | Run was started with 1.0% Armid added to the shell formulation but capsules would not form. Shell without Armid was added to give 0.2% Armid. Capsules formed well. Some multicore. Polyox used. |
| 1-18A1 | Sunoco 4412 wax Piccolyte S-115 Gulf P.E. 2205 | 50 45 5 | 64.8 | 2.21 | 3000 | 70 | Capsules large and irregular. Many multicore. No empties. Polyox used. |
| 1-18A2 | Same as 1-18A1 | | 64.8 | 2.21 | 4000 | 64-71 | Capsules looked good except many are multicore. No empties. Polyox used. |
| 1-18B | Sunoco 4412 wax Piccolyte S-115 Epolene C-16 | 40 40 20 | 64.8 | 2.21 | 3600 | 79-80 | Many elongated and multicore capsules. No empties. At 3000mL/min carrier, capsules large and irregular. |

¹ Indicates sample submitted.

* Runs using basic formulation.

** Runs using polyethylene with paraffin and Piccolyte.

*** Runs using formulation of choice.

Table I (Cont'd)

| Run No. (SRI No.) ¹ | Shell Composition | % | Estimated Payload wt. % | Estimated Production kg/hr | Carrier Rate mL/min | Carrier Temp. °C | Comment |
|-----------------------------------|--|---------------------|-------------------------------|----------------------------------|---------------------------|------------------------|---|
| 1-20A1** | Sunoco 4412 wax Piccolyte S-115 Epolene C-16 | 55 30 15 | 64.8 | 2.21 | 3000 | 82 | 0.5% Polyox WSR-35 was used in this and all other runs shown. Many multicore capsules. Almost no empty capsules after overnight drying. |
| 1-20A2** | Same as 1-20A1 | | 64.8 | 2.21 | 2800 | 83 | More large multicore capsules than 1-20A1. |
| 1-20B1 | Sunoco 4412 Piccolyte S-115 Epolene C-16 Polybutene 128 | 50 30 15 5 | 64.8 | 2.21 | 3000 | 81 | No empty capsules. |
| 1-20B2 | Same as 1-20B1 | | 64.8 | 2.21 | 3300 | 79 | Capsules appear better than 1-20B1. Less large multicore capsules. |
| 1-23A* | Sunoco 4412 Piccolyte S-115 Polybutene 128 | 40 47 13 | 64.8 | 2.21 | 3000 | 65-66 | No sticking or plugging. Some blocking in collection vessel. Large number of satellites. No empties on production. |
| 1-23B* | Same as 1-23A | | 64.8 | 2.21 | 3400 | 63-64 | Capsules look better than 1-23A. Block slightly more on collection. |
| 1-24A | Sunoco 4412 Piccolyte S-115 Elvax 210 | 40 40 20 | 64.8 | 2.21 | 3560 | 77-78 | Capsules all filled but poorly formed. Off center and multicored. |
| 1-24B | Sunoco 4412 Piccolyte S-115 Epolene C-16 Polybutene 128 | 35 40 20 5 | 64.8 | 2.21 | 3000 | 79 | Capsules not formed. Stringing and elongated particles. Heat control at maximum. No sample. |
| 1-27A** | Sunoco 4412 Piccolyte S-115 Epolene C-16 | 50 35 15 | 64.8 | 2.21 | 3520 | 70 | New shell and fill tubes installed. Outer orifice 0.155" (same as old), inner tube 0.042" OD x 0.28" ID (old was 0.050" OD x 0.031" ID). Made well when aligned. Small sample. Some agglomerates and multicore. |
| 1-27B1 | Sunoco 4412 Piccolyte S-115 Epolene C-16 Polybutene 128 | 45 35 15 5 | 64.8 | 2.21 | 3520 | 74 | Made well. Some multicore and some empties after drying overnight. |
| 1-27B2 | Same as 1-27B1 | | 64.8 | 2.21 | 3600 | 71-72 | Not as well formed as 1-27B1. More small satellites than 1-27A. Some multicore. Some empty after drying. |
| 1-27C** | Sunoco 4412 Piccolyte S-115 Epolene C-16 | 50 40 10 | 64.8 | 2.21 | 3520 | 71-74 | Made well. Some multicore. Many empty on drying. |
| 1-28A1** | Sunoco 4412 Piccolyte S-115 Epolene C-16 | 45 45 10 | 64.8 | 2.21 | >3600 | 72-76 | Capsules filled on collection. Capsules not as good when temperature over 76°C. |
| 1-28A2** | Same as 1-28A1 | | 64.8 | 2.21 | 3600 | 75 | Capsules look good. |
| 1-28A3** | Same as 1-28A1 | | 64.8 | 2.21 | 3280 | 75 | Capsules look good. |
| 1-28B1** | Sunoco 4412 Piccolyte S-115 Epolene C-16 | 48 40 12 | 64.8 | 2.21 | 3200 | 69-75 | Capsules filled on collection except a few large empty capsules. |

¹ Indicates sample submitted.

* Runs using basic formulation.

** Runs using polyethylene with paraffin and Piccolyte.

*** Runs using formulation of choice.

Table I (Cont'd)

| Run No. (SWR) No.) ¹ | Shell Composition | % | Estimated Payload wt. % | Estimated Production kg/hr | Carrier Rate mL/min | Carrier Temp. °C | Comment |
|------------------------------------|---|---------------------|-------------------------------|----------------------------------|---------------------------|------------------------|---|
| 1-28B2** | Same as 1-28B1 | | 64.8 | 2.21 | 3320 | 73-74 | Capsules look good. |
| 1-28C | Sunoco 4412 Piccolyte S-115 Polybutene 128 Armid HT added to above 0.3% | 40 47 13 | 64.8 | 2.21 | 3320 | 63-65 | Made well. Many satellites. |
| 1-32A1** | Sunoco 4412 Piccolyte S-115 Epolene C-16 | 40 47 13 | 64.8 | 2.21 | 3200 | 70-73 | New inner tube 0.064" OD x 0.043" ID. Capsules look good. 70°C is too low. |
| 1-32A2** | Same as 1-32A1 | | 64.8 | 2.21 | 3600 | 75 | Made well. Appear to be more single capsules than with other nozzle. |
| 1-32B1 | Shellwax 700 Piccolyte S-115 Sunoco 4412 | 20 45 35 | 64.8 | 2.21 | 3600 | 72-76 | Below 72°C shell clumps and strings. Flow rate and temperature appear critical. |
| 1-32B2 | Same as 1-32B1 | | 64.8 | 2.21 | 3600 | 75-76 | Some elongation. Capsules filled on collection but many empty after drying overnight. |
| 1-35A1 | Sunoco 4412 Piccolyte S-115 Epolene C-16 Polybutene 128 | 40 45 10 5 | 64.8 | 2.21 | 2900-3150 | 76-79 | Made well. |
| 1-35A2 | Same as 1-35A1 | | 64.8 | 2.21 | 3300 | 76-79 | Made well. Almost no empty capsules after drying overnight. |
| 1-36 | Same as 1-35A1 except 0.5% Aluminum Stearate added | | 64.8 | 2.21 | 3500-3600 | 77 | Some capsules elongated. Change in carrier rate and temperature not helpful. No empties after drying overnight. |
| 1-36A* | Sunoco 4412 Piccolyte S-115 Polybutene 128 | 40 47 13 | 64.8 | 2.21 | 3360 | 60-64 | 0.5% Polyox WSR-35 was used in the carrier in this and all other runs shown. Fill water contained 0.005% blue dye. Capsules formed well. Some blocking occurred on collection screen. This became bad at ca. 64°C carrier temperature. |
| 1-38B* (7-059) (7-060) | Same as 1-38A | | 65.8 | 2.19 | 3360 | 58-62.5 | Shell reservoir temperature reduced from 74°C to 71°C. Fill reservoir temperature reduced from 67°C to 58°C. Ran 1 hour and 40 minutes without problems. Very little blocking. |
| 1-38C* | Same as 1-38A | | 65.2 | 3.45 | 3360 | 60-65 | Made well. No more blocking than at lower production rate. Capsules were larger. |
| 1-40 | Same as 1-38A with 0.3% Armid HT added | | 65.2 | 3.45 | 3360 | 59-63 | Slight reduction in capsule size from 1-38C capsules. Little or no blocking. |
| 1-42A* | Same as 1-38A | | 65.4 | 9.1 | 3400-4400 | 61-65 | Shell reservoir-71°C, Fill-58°C. Capsules large and stuck together on screen. Cap- sules broke apart with some damage at 61°C but stuck at 65°C. No sample. |

¹ Indicates sample submitted.

- * Runs using basic formulation.
- ** Runs using polyethylene with paraffin and Piccolyte.
- *** Runs using formulation of choice.

Table I (Cont'd)

| Run No. (SwRI No.) ¹ | Shell Composition | % | Estimated Payload wt.-% | Estimated Production kg/hr | Carrier Rate mL/min | Carrier Temp. °C | Comment |
|------------------------------------|--|---------------------|-------------------------------|----------------------------------|---------------------------|------------------------|---|
| 1-428* | Same as 1-38A | | 65.4 | 9.1 | 4400 | 63 | Shell reservoir temperature reduced to 65.0°C. Fill reservoir reduced to 50°C. Still blocked badly on screen. A small sample caught in a beaker and poured on the screen did not block. |
| 1-43A* | Same as 1-38A | | 65.4 | 9.1 | 4000 | 59-65 | A five-gallon surge tank was installed before the capsule collection screen. This stopped blocking on screen. Fill reservoir temperature was 42°C. Capsules were large and poorly formed. |
| 1-43B* | Same as 1-38A | | 65.4 | 9.1 | 4300 | 60-64 | Capsules smaller and better than 1-43A but still large and many empty capsules. |
| 1-43C | Same as 1-38A but 0.3% Armid HT added | | 64.0 | 8.7 | 5000 | 60-64 | Capsules smaller than 1-43B. Most capsules broke until fill temperature raised to 60°C and fill rate reduced. Still some broken capsules, but much better. Many satellites. |
| 1-45* | Sunoco 4412 Piccolyte S-115 AC-617 polyethylene | 40 45 15 | 64.0 | 8.7 | 5350 | 70 | Shell reservoir was 87.8°C. Fill reservoir 55°C. Capsules not broken but did not form well. Many solid particles and wide size range and payload variation. |
| 1-46 (7-061) | Sunoco 4412 Piccolyte S-115 Epolene C-16 Polybutene 128 | 50 30 15 5 | 65.0 | 2.28 | 3000 | 78-79 | Inner nozzle 0.064" OD x 0.043" ID. 0.5% Polyox WSR-35 in carrier water. Shell reservoir 88°C. Fill reservoir 55°C raised to 65°C. 0.005% blue dye in fill. Capsule size was a little larger and range wider than desired. Temperature at top of controller range. Capsules looked good. 1.9kg shipped. |
| 1-47A* | Sunoco 4412 Piccolyte S-115 Polybutene 128 | 40 47 13 | 63.8 | 2.39 | 3360-3400 | 60-64 | Shell reservoir 71°C. Fill - 58-59°C. Capsule size was large and many satellites were formed. |
| 1-47B* | Same as 1-47A | | 63.8 | 2.39 | 3200 | 63-65 | Capsules much better looking than 1-47A. Conditions same. Shell used contained shell which had been heated before and may have been more oxidized. |
| 1-49A* | Same as 1-47A | | 65.5 | 9.08 | 5580 | 64 | Inner nozzle 0.081" OD x 0.062" ID. 0.2% Polyox WSR-205 in carrier water. Shell reservoir 68°C; Fill 60°C. Five-gallon carrier surge tank used, but capsules stuck some on recovery. Capsules looked good but large. |
| 1-49B* | Same as 1-47A | | 65.5 | 9.08 | 5580 | 62 | About the same as 1-49A. Capsules still stick some. |
| 1-49C* | Same as 1-47A | | 65.5 | 9.08 | 5820-6000 | 60-62 | Conditions same. Capsules smaller and look good but too hot and agglomerated in tank and on screen. |

¹ Indicates sample submitted.

* Runs using basic formulation.

** Runs using polyethylene with paraffin and Piccolyte.

*** Runs using formulation of choice.

Table I (Cont'd)

| Run No. (SwRI No.) ¹ | Shell Composition | % | Estimated Payload wt. % | Estimated Production kg/hr | Carrier Rate mL/min | Carrier Temp. °C | Comment |
|------------------------------------|---|-----------------------------|-------------------------------|----------------------------------|---------------------------|------------------------|--|
| 1-50A* | Same as 1-47A | | 66.5 | 9.08 | 6300 | 59.5 | Shell - 65.5°C; Fill - 53°C. Tank - 40-41°C. Capsules look good but too hot-stick badly on collection. |
| 1-50B* | Same as 1-47A | | 65.5 | 9.08 | 6000 | 58 | Fill - 50°C; Tank - 40°C. A little better, but capsules still stick on collection. |
| 1-51A* | Same as 1-47A | | 65.5 | 9.08 | 6000 | 59-60 | New batch of shell (heated 48 hr at 70°C). Other conditions same. Stuck about as 1-50B. |
| 1-51B1 | Same as 1-47A but 0.3% Aramid HT added | | 65.5 | 9.08 | 6000 | 59-61 | Look good but stick on collection. |
| 1-51B2 | Same as 1-51B1 | | 65.5 | 9.08 | 6000 | 58-59 | Do not appear to stick as badly as 1-51B1. |
| 1-51C1 | Sunoco 4412 Piccolyte S-115 Polybutene 128 Epoilene C-16 | 36.4 42.7 11.8 9.1 | 65.5 | 9.08 | 5400 | 65 | Shell - 88°C; Fill - 67°C; Tank - 40.5°C. Many large capsules, and capsules stick on collection. |
| 1-51C2 | Same as 1-51C1 | | 65.5 | 9.08 | 6000 | 65 | Capsules smaller but still stick together badly. |
| 1-53A* | Sunoco 4412 Piccolyte S-115 Polybutene 128 | 40 47 13 | 65.5 | 9.08 | 6000 | 58-60 | Shell - 74°C; Fill - 47.5°C. 0.2% Polyox 205 in fill as well as in carrier water. Made well but stick and still large. |
| 1-53B* | Same as 1-53A | | 65.5 | 9.08 | 5700 | 57 | Shell - 71°C; Fill - 47°C. Other conditions same. Still stick some. |
| 1-53C* | Same as 1-53A | | 65.5 | 9.08 | 5640 | 56.5 | Shell - 71°C; Fill - 45°C. Other conditions same. Capsules form well. Still stick together. |
| 1-53D* | Same as 1-53A | | 65.5 | 9.08 | 5640 | 56-56.5 | Shell - 68°C; Fill - 44°C. Other conditions same. Still stick some. Some capsules have tails. |
| 1-55A | Sunoco 4412 Piccolyte S-115 Shellwax 700 Polybutene 128 | 40 45 5 10 | 65.5 | 9.08 | 5700 | 61 | Conditions same as 1-53A except Shell reservoir - 71°C; Fill - 45°C. Trace of blue dye. Strung and plugged system. No capsules. |
| 1-55B | Same as 1-55A | | 65.5 | 9.08 | 6000 | 66 | Same conditions as 1-55A except Shell - 79.4°C; Fill - 65°C. Made good capsules. Very little sticking. |
| 1-55C | Same as 1-55A | | 65.5 | 9.08 | 6000 | 63-65 | Same conditions as 1-55B but variation in carrier temperature. A little less sticking than 1-55B. |
| 1-56A*** | Sunoco 5512 wax Piccolyte S-115 Polybutene 128 | 45 45 10 | 65.5 | 9.08 | 6300 | 65 | Same conditions as 1-55B but 0.1% Polyox 205 in carrier. Fill - 55°C. Temperature of carrier water in surge tank was 40°C. Capsules did not stick in tank or collection screen. Look good and are not tacky. |

¹ Indicates sample submitted.

* Runs using basic formulation.

** Runs using polyethylene with paraffin and Piccolyte.

*** Runs using formulation of choice.

Table I (Cont'd)

| Run No. (SwRI No.) ¹ | Shell Composition | % | Estimated Paraffin wt.-% | Estimated Production kg/hr | Carrier Rate mL/min | Carrier Temp. °C | Comment |
|------------------------------------|---|---------------------------|--------------------------------|----------------------------------|---------------------------|------------------------|---|
| 1-55B*** | Same as 1-56A | | 65.5 | 9.08 | 7000 | 63-64 | Same conditions as 1-56A. Capsules smaller and more uniform than 1-56A. No sticking. |
| 1-56C | Sunoco 4412 Piccolyte S-115 Epolene C-16 Polybutene 128 | 40 40 10 10 | 65.5 | 9.08 | 7000 | 63-64 | Same conditions as 1-56B except Shell - 82°C; Fill - 63°C. Capsules made well but stuck together in surge tank. Broke apart easily when cold. Many elongated capsules. |
| 2-1 | Sunoco 5512 Piccolyte S-115 Epolene C-16 Polybutene 128 | 45 40 5 10 | 65.5 | 9.08 | 7300-6500 | 62-65 | Capsule fill was deionized water with 0.005% Pyram deep blue W-13 dye and 0.1% Polyox WSR-205 added. Fill reservoir was 53°C; shell was 82°C. Capsules did not form well-tails. (No sample. (0.1% Polyox WSR-205 in carrier.) |
| 2-2 | Sunoco 4412 Sunoco 5512 Piccolyte S-115 Epolene C-16 Polybutene 128 | 32 10 42 6 10 | 65.5 | 9.08 | 7000 | 64 | Same fill and conditions as 2-1. Temperature of collection tank was 42°C. Some clustering in tank but no real sticking. |
| 2-4A*** | Sunoco 5512 Piccolyte S-115 Polybutene 128 | 45 45 10 | 65.5 | 9.08 | 7000 | 64.5 | Same fill. Shell - 88°C; Fill - 60°C. About 0.06% Polyox WSR-301 in carrier. Temperature of collection tank was 43.9°C. Capsules stick very slightly in collection sieve. |
| 2-4B*** | Same as 2-4A | | 65.5 | 9.08 | 7300 | 64 | Other conditions same as 2-4A. Tank was 44.4°C. Sticking still slight. Capsules look good. |
| 2-4C*** | Same as 2-4A | | 65.5 | 9.08 | 7500 | 64 | Same as 2-4B. Capsules look good. |
| 2-6A*** | Same as 2-4A | | 65.5 | 9.08 | 7500 | 64 | Same as 2-4A but fill - 62°C, carrier 0.04% Polyox WSR-301, collection tank - 43.5°C. Capsules not as good as 2-4C. |
| 2-6B*** | Same as 2-4A | | 65.5 | 9.08 | 7500 | 64 | Same as 2-6A but 0.1% Polyox WSR-N-12K with 0.17% isopropanol in carrier instead of WSR-301. Capsules look about like 2-4C. Collection tank - 42.4°C. |
| 2-8*** (7-079) | Sunoco 5512 Piccolyte S-115 Polybutene 128 | 45 45 10 | 65.0 | 4.8 | 6700-7000 | 63-65 | Fill was deionized water with 0.1% Polyox WSR-205 and 0.005% blue dye added. Carrier water contained 0.05% Polyox WSR-301. Fill was 60°C; Shell was 93°C. Collection tank was 46°C. Slight sticking on collection, but capsules break up easily cold. Run was about 3 hrs. 25 lbs of capsules were shipped. |
| 2-9A | Boler wax 1435 Piccolyte S-115 Polybutene 128 | 45 45 10 | 65.0 | 4.8 | 6500 | 64-65 | Same conditions as Run 2-8. Capsules stuck together more than 2-8 capsules. Some clumps did not break up easily when cold. Most did part easily. |

¹ Indicates sample submitted.

- * Runs using basic formulation.
- ** Runs using polyethylene with paraffin and Piccolyte.
- *** Runs using formulation of choice.

Table I (Cont'd)

| Run No. (SWR) No. ¹ | Shell Composition | % wt. | Estimated Payload wt. % | Estimated Production kg/hr | Carrier Rate ml/min | Carrier Temp. °C | Comment |
|-----------------------------------|--|--------------------------|-------------------------------|----------------------------------|---------------------------|------------------------|---|
| 2-981 | Boler wax 1426 Piccolyte S-115 Polybutene 128 | 45 45 10 | 65.0 | 4.8 | 6500 | 63-64 | Same conditions as Run 2-2. Capsules stuck together more than 2-9A, but still not bad. |
| 2-982 | Same as 1-981 | | 65.0 | 4.8 | 6700 | 61-62 | Same conditions as Run 2-8 except: Shell was 88°C; Fill was 57°C; and collection tank was 44°C. Strings and clumps formed with no good capsules. No sample was made. |
| 2-983 | Same as 2-981 | | 65.0 | 4.8 | 6100 | 63-64 | Same conditions as Run 2-8 except: collection tank was 43-44°C. Capsules formed well, but sticking occurred. |
| 2-12A | Boler wax 1426 Piccolyte S-115 | 53 47 | 65.0 | 4.8 | 6200 | 62-65 | Same conditions as Run 2-8 except: Fill was 58°C and collection tank was 44-45°C. At 62 to 63°C carrier temperature, some empty capsules were produced. Some sticking occurred at 64-65°C but not bad. |
| 2-128** | Boler wax 1426 Piccolyte S-115 Epolene C-16 | 50 45 5 | 65.0 | 4.8 | 4500 | 75 | Same conditions as Run 2-8 but collection tank was 44°C. Good capsules would not form under 73°C carrier temperature. No sticking occurred with these capsules. |
| 2-14A** | Sunoco 4412 Boler wax 1426 Piccolyte S-115 Epolene C-16 | 30 20 45 5 | 65.0 | 4.8 | 5300 | 67-68 | Same conditions as Run 2-8 but collection tank was 43°C. Capsules formed with no sticking. |
| 2-14B | Sunoco 4412 Boler wax 1426 Piccolyte S-115 Epolene C-16 Polybutene 128 | 25 20 45 5 5 | 65.0 | 4.8 | 5300 | 68 | Same conditions as 2-14A. Slight clumping in collection bath. Many empties after drying overnight. |
| 2-20A | Boler wax 1426 Piccolyte S-115 Polybutene 128 | 45 45 10 | 65.6 | 9.17 | 7500 | 65 | Fill was deionized water with 0.05% Polyox WSR-205 and 0.005% blue dye added. Shell reservoir 93°C; Fill - 63°C. Booster pump on cooling water line reduced collection tank temperature to 40°C. No capsule sticking at this temperature. |
| 2-20B | Same as 2-20A | | 65.6 | 9.17 | 8000 | 64 | Conditions same as 2-20A. Carrier pump caused vibration but good capsules with no sticking. |
| 2-21 | Same as 2-20A | | 65.6 | 9.17 | 8000 | 64-65 | Conditions same as 2-20A. Carrier pump vibration reduced. Capsules smaller and more uniform. |
| 2-22A | Same as 2-20A | | 69.2 | 10.22 | 8000 | 64 | Conditions same as 2-20A except Fill reservoir 62°C. Capsules good until carrier water dropped to 60.5°C and tails formed on capsules. |

¹ Indicates sample submitted.

* Runs using basic formulation.

** Runs using polyethylene with paraffin and Piccolyte.

*** Runs using formulation of choice.

Table I (Cont'd)

| Run No. (SwRI No.) ¹ | Shell Composition | % | Estimated Payload wt. % | Estimated Production kg/hr | Carrier Rate mL/min | Carrier Temp. °C | Comment |
|------------------------------------|--|--------------------------|-------------------------------|----------------------------------|---------------------------|------------------------|---|
| 2-22C* | Sunoco 4412 wax Piccolyte S-115 Polybutene 128 | 40 47 13 | 69.2 | 10.22 | 7500 | 59-62 | Conditions same as 2-22A. Chiller used. Collection tank was 30-32°C. Capsules made well with no sticking. Heating system did not maintain temperature at 62°C. |
| 2-24A* | Same as 2-22C | | 69.2 | 10.22 | 7400 | 60-61 | Conditions same as 2-22A. Chiller used. Capsules made well with no sticking. |
| 2-24B*** | Sunoco 5512 (Ross) Piccolyte S-115 Polybutene 128 | 45 45 10 | 69.2 | 10.22 | 8000 | 65 | Conditions same as 2-22A. No chiller. No collection tank. Carrier temperature out - 39°C. Temperature steady. Made well. No sticking. |
| 2-24C*** | Same as 2-24B | | 65.6 | 9.16 | 8100 | 65-66 | Conditions same as 2-24B. Made well with no problems. |
| 2-26 | Sunoco 5512 Piccolyte S-115 Polybutene 128 Carnauba wax Epolene C-16 | 43 42 10 2 3 | 65.6 | 9.16 | 7000 | 66-68 | Conditions same as 2-24B. Made well. Heating system would not maintain temperature at 68°C. |
| 2-29A*** | Sunoco 5512 Piccolyte S-115 Polybutene 128 | 45 45 10 | 65.6 | 9.16 | 7900 | 65-66 | Fill was water with 0.05% Polyox WSR-205 and 0.005% blue dye. Fill temperature was 62°C, Shell temperature 90.5°C. Capsule recovery was 8.54 kg/hr. Capsules were more uniform in size than 2-29B below with fewer elongated. |
| 2-29B*** | Same as 2-29A | | 65.6 | 9.16 | 7900 | 65-66 | Run was same as 2-29A, but with no Polyox in the fill. Capsule recovery was 8.4 kg/hr. |
| 2-31A*** (7-080) | Same as 2-29A | | 65.6 | 9.16 | 8000 | 64-65 | Conditions same as 2-29A, but 0.03% Pylam Deep Blue W-13 dye in the fill. Sample shipped was 1000u to 2300u. |
| 2-31B*** (7-081) | Same as 2-29A | | 65.6 | 9.16 | 8000 | 64-65 | Conditions same as 2-29A, but 0.05% Pylaklor Brilliant Red S-363 in the fill. Sample shipped was 1000u to 2300u. |
| 2-32*** (7-082) | Same as 2-29A | | 69.3 | 10.25 | 8000 | 64 | Conditions same as 2-29A except higher fill rate. Actual capsule recovery was 9.43 kg/hr. |
| 2-35*** | Sunoco 5512 Piccolyte S-115 Polybutene 128 | 45 45 10 | 67.8 | 9.78 | 8000 | 62-68 | Encapsulation system was installed in new building with new steam supply. Fill for this and following runs was deionized water with 0.05% Polyox WSR-205 and 0.005% blue dye. Carrier fluid was water with 0.05% Polyox WSR-301. Shell reservoir was 93°C; fill reservoir 60°C. Carrier temperature variation was excessive with slight sticking of capsules at 68°C. Capsules were of good appearance. |
| 2-36A*** | Same as 2-35 | | 67.8 | 9.78 | 8000-7600 | 62-68 | Conditions same as 2-35. Cooling water flow restricted and some variation in carrier rate and temperature. Some slight capsule sticking when carrier about 68°C, but capsules appear good and undamaged. |

¹ Indicates sample submitted.

- * Runs using basic formulation.
- ** Runs using polyethylene with paraffin and Piccolyte.
- *** Runs using formulation of choice.

Table I (Cont'd)

| Run No. (SwRI #p.) ¹ | Shell Composition | % | Estimated Payload wt. % | Estimated Production kg/hr | Carrier Rate mL/min | Carrier Temp. °C | Comment |
|------------------------------------|--|----------------|-------------------------------|----------------------------------|---------------------------|------------------------|--|
| 2-36B*** | Same as 2-35 with 0.02% BHT added | | 67.8 | 9.78 | 8000-7600 | 62-68 | Conditions same as 2-36A. No effect of the butylated hydroxytoluene antioxidant noted. |
| 2-37A*** | Same as 2-36B | | 67.8 | 9.78 | 8000 | 61-64 | Inner nozzle size increased from 0.081" OD x 0.062" ID to 0.095" OD x 0.077" ID. Outer nozzle orifice still 0.155". Cooling system was cleaned and flow increased to 10.5 liters per minute. Other conditions were not changed. Carrier fluid at collection was 36°C. When carrier temperature dropped to 61°C, stringing occurred. |
| 2-37B*** | Same as 2-36B | | 67.8 | 9.78 | 8000 | 64-66 | Cooling water flow was cut to about 9.1 liters per min. Other conditions were unchanged. Carrier fluid at collection was 37°C, and no sticking of capsules occurred. Capsules looked good. |
| 2-38*** | Same as 2-36B | | 67.8 | 9.78 | 8000 | 66-67.5 | Conditions same as 2-37A. Temperature at collection 39°C. No sticking. Capsule collection rate was 9.04 kg/hr. Capsules looked good. |
| 2-39A*** | Same as 2-35 | | 65.2 | 11.32 | 8200 | 66-67 | Conditions same as 2-37A but cooling water 6.6 liters per min. Capsule collection rate was 10.57 kg/hr. Capsules looked good. |
| 2-39B*** | Same as 2-35 | | 65.2 | 11.32 | 8800 | 67 | Conditions same as 2-37A. Temperature at collection 40°C. No sticking. Capsules look good. Capsule collection rate was 10.59 kg/hr. |
| 2-41A*** | Same as 2-35 | | 65.0 | 12.70 | 9300 | 61-63 | Conditions same as 2-37A, but cooling water 7.5 liters/min and temperature at collection was 40°C. Collection rate was 12.03 kg/hr. Capsules looked good. |
| 2-41B*** | Same as 2-35 | | 65.0 | 12.70 | 9300 | 65-66 | Conditions not changed. Temperature at collection was 41.7° to 42.2°C. Slight sticking occurred, but capsules parted without apparent damage and looked good. |
| 2-42A*** | Sunoco 5512 wax Piccolyte S-115 Polybutene 128 | 45 45 10 | 68.9 | 14.00 | 9800 | 61-66 | Fill was deionized water with 0.05% Polyox WSR-205 and 0.005% blue dye added. Carrier liquid was water with 0.05% Polyox WSR-301 added. Collection temperature was 42°C. Shell reservoir was 93°C; Fill reservoir was 60°C. Outer orifice was 0.155". Inner nozzle was 0.095" OD x 0.077" ID. Capsule collection rate was 13.4 kg/hr. Some badly formed capsules when temperature is below 62°C but sample of good appearance. |
| 2-42B*** | Same as 2-42A | | 68.9 | 14.00 | 9800 | 64-67 | Conditions same as 2-42A except temperature. Capsules were good. |

¹ Indicates sample submitted.

- * Runs using basic formulation.
- ** Runs using polyethylene with paraffin and Piccolyte.
- *** Runs using formulation of choice.

Table I (Cont'd)

| Run No. (Su.R. No.) ¹ | Shell Composition | % | Estimated Payload wt. % | Estimated Production kg/hr | Carrier Rate mL/min | Carrier Temp. °C | Comment |
|-------------------------------------|--|----------------|-------------------------------|----------------------------------|---------------------------|------------------------|---|
| 2-44A*** (7-083) | Same as 2-42A | | 65.5 | 8.62 | 8500 | 68 | Fill reservoir 65°C. Fill was deionized water with 0.05% Polyox WSR-205 and 0.06% Brilliant Red dye S-353. Collection temp. was 40°C. Other conditions not changed. Capsules were good. |
| 2-44B*** (7-084) | Same as 2-42A | | 65.5 | 8.62 | 8500 | 68 | Red dye was increased to 0.11% of fill. Other conditions were the same as 2-44A. Capsules were good. |
| 2-45*** | Same as 2-42A | | 65.1 | 16.58 | 11000 | 66-70 | Fill was a mixture of 2-42A and 2-44B fill. Fill temperature was 60°C. Collection temperature was 43°C. Other conditions were not changed. Made all right but change in size distribution and more satellites formed. |
| 2-46*** | Same as 2-42A | | 65.3 | 18.28 | 11000 | 67-69 | Fill same as 2-42A. Fill temperature 65°C. Other conditions not changed. Made about as 2-45. Fill pump apparently did not pump to set rate when under pressure. |
| 2-47A*** | Same as 2-42A | | 65.1 | 22.62 | 11000 | 68-72 | Fill same as 2-42A. Other conditions not changed. Change in size distribution and many satellites. Fill pump not pumping set rate since both capsule collection rate and payload low. Capsules not well formed. |
| 2-47B*** | Same as 2-42A | | 65.1 | 22.62 | 11000 | 64-70 | Run was the same as 2-47A except carrier temperature. Capsules were about the same. |
| 2-47C*** | Same as 2-42A | | 65.1 | 22.62 | 11000 | 65-67 | Same as 2-47B. |
| 2-49A*** | Sunoco 5512 Piccolyte S-115 Polybutene 126 | 45 45 10 | 70.9 | 27.2 | 11000 | 65-66 | Fill was deionized water with 0.05% Polyox WSR-205 and 0.005% blue dye added. Carrier fluid was water with 0.05% Polyox WSR-301 added. Outer orifice was 0.155". Inner nozzle was 0.095" OD x 0.077" ID. Shell reservoir 60°C. Vibration of carrier pump was reduced. Fill pump rate was increased to compensate for loss under pressure. Capsules produced were poorly formed with many empty. |
| 2-49B*** | Same as 2-49A | | 70.9 | 27.2 | 10000 | 65-67 | Other conditions same as 2-49A. Capsules of poor quality. |
| 2-50A*** | Same as 2-49A | | 65.2 | 22.7 | 10500 | 67-68 | Other conditions same as 2-49A, but new fill pump used. Capsules better than 2-49 but still some empty. Collection temperature was high (42°C). |
| 2-50B*** | Same as 2-49A | | 65.2 | 22.7 | 11000 | 68 | Capsules of poor quality. Collection temperature 42°C. Some agglomeration. |
| 2-50C*** | Same as 2-49A | | 65.2 | 22.7 | 10500 | 65 | Chiller was used to reduce collection temperature to 32°C. No agglomeration. Some empty capsules and multicore capsules. |

¹ Indicates sample submitted.

- * Runs using basic formulation.
- ** Runs using polyethylene with paraffin and Piccolyte.
- *** Runs using formulation of choice.

Table I (Cont'd)

| Run No. (SWRI No.) ¹ | Shell Composition | Estimated Payload wt. % | Estimated Production kg/hr | Carrier Rate mL/min | Carrier Temp. °C | Comment |
|------------------------------------|-------------------|-------------------------------|----------------------------------|---------------------------|------------------------|---|
| 2-51A*** | Same as 2-49A | 65.4 | 22.7 | 11300 | 67-68 | Conditions same as 2-50C but nozzle was realigned and moved from about 1/4" back of orifice to 1/8" back of orifice. Capsules look better than 2-50C, but some small empty capsules and some with tails. |
| 2-51B*** | Same as 2-49A | 65.4 | 22.7 | 10600 | 64-66 | Conditions same as 2-51A. Capsules were larger. |
| 2-52*** | Same as 2-49A | 65.4 | 22.7 | 11000 | 68-73 | Conditions not changed. Capsules look better but a few still have tails. |
| 2-53A*** | Same as 2-49A | 65.4 | 22.7 | 10700 | 66-70 | Fill reservoir was 70°C and shell reservoir 95°C. Capsules may look slightly better than 2-52. |
| 2-53B*** | Same as 2-49A | 65.4 | 22.7 | 10700 | 68-70 | Other conditions same as 2-53A. Capsules about the same. |
| 2-53C*** | Same as 2-49A | 65.4 | 22.7 | 10700 | 72-75 | Other conditions same as 2-53A. Capsules about the same. |
| 2-53D*** | Same as 2-49A | 65.4 | 22.7 | 10500 | 76-77 | Other conditions same as 2-53A but shell reservoir 96°C and fill 74°C. Capsules better than 2-53A, B and C. |
| 2-53E*** | Same as 2-49A | 65.4 | 22.7 | 10300 | 76-77 | Other conditions same as 2-53D. Capsules not quite as good as 2-53D. |
| 3-1A*** | Same as 2-49A | 65.4 | 22.7 | 10500 | 79-82 | Other conditions same as 2-53E. Capsules appear better than 2-53D. |
| 3-1B*** | Same as 2-49A | 65.2 | 22.7 | 10100 | 80-83 | Other conditions not changed. A filter was installed to prevent satellites from accumulating and melting in the heat exchanger. Capsules were about the same quality as 3-1A. Material balance and payload indicate that shell pump is not pumping at set rate. |
| 3-4A*** | Same as 2-49A | 65.9 | 18.6 | 10000 | 78-80 | Shell transmission was changed. Shell reservoir was 96°C, fill 73°C. A few empty capsules were present but payload was near that estimated. |
| 3-4B*** | Same as 2-49A | 65.9 | 18.6 | 9800 | 75 | Conditions not changed. Capsules did not look as good as 3-4A. |
| 3-5*** | Same as 2-49A | 65.9 | 18.6 | 9900 | 78-80 | Fill nozzle moved from 1/8" back of orifice to 1/16" back of orifice. Capsules may be slightly better in appearance but still some empty. |
| 3-6A*** | Same as 2-49A | 65.7 | 15.4 | 10000 | 75-76 | Conditions not changed. Some empty capsules. |
| 3-6B*** | Same as 2-49A | 65.7 | 15.4 | 9600 | 75-77 | Conditions not changed. Capsules better formed. |
| 3-8A*** | Same as 2-49A | 66.6 | 14.3 | 9500 | 75-80 | New transmission pumping system and heating system for both fill and shell. Capsules look all right but a few empties. Ran ca. 1-1/2 hours to prepare sample. |
| 3-9*** | Same as 2-49A | 67.4 | 12.0 | 10500 | 75-80 | Capsules for sample. Ran 2 hours and 15 minutes total. |

¹ Indicates sample submitted.

- * Runs using basic formulation.
- ** Runs using polyethylene with paraffin and Piccolyte.
- *** Runs using formulation of choice.

NOTES TO TABLE I

1. Carrier Flow. The carrier fluid is pumped from a reservoir at the collection end of the system through a heating system, around the nozzle where shell and fill are injected into it for capsule formation, through a convergence section, followed by a jacketed tube where the fluid is cooled to solidify the capsule shell. The carrier fluid and capsules are separated when the capsules are collected on a screen, and the carrier fluid is returned to the reservoir. Carrier flow rate has a major effect on the heating and cooling requirements and is a major factor in determining capsule size. Turbulence in the carrier stream or fluctuation in rate are factors in capsule uniformity.
2. Carrier Fluid. The usual carrier fluid used in the system for a non-aqueous hot melt shell is tap water, but modifiers can be used to reduce turbulence and retard shell stickiness.
3. Carrier Temperature. Carrier temperature is a major factor in capsule formation and capsule size. The temperature of the carrier at collection affects blocking and sticking which may result in holes in the capsules.
4. Nozzle Size and Configuration. Nozzle size and configuration has an effect on the size of the capsules. Turbulence results when shell, fill and carrier streams differ in velocity, and the turbulence affects capsule quality. The centering of the tubes and the location of the nozzle orifices with reference to each other and to the convergence in the tube are factors in capsule formation.
5. Shell and Fill Rates. Shell and fill rates determine payload and production rate. A steady, nonphasing, positive flow is needed to prevent differences in payload and size. Shell and fill pumps used were gear pumps, immersed in an oil bath with shell and fill reservoirs to provide temperature control.
6. Shell and Fill Temperatures. Shell and fill temperatures may be varied above or below the carrier fluid temperature with an effect on capsule formation.
7. Cooling Fluid Flow Rate and Temperature. Cooling fluid flow rate and temperature control temperature gradient across the jacketed tube.
8. Capsule Shell Formulation. Viscoelastic properties and hardening characteristics require system adjustment to form satisfactory capsules of the desired size.

TABLE II. CAPSULE SAMPLE ANALYSIS

| Run No. (SwRI No.) ⁽¹⁾ | Payload wt. % | Burst Strength, g | Size Distribution - % | | | Water Loss Rate - % Original Fill | | | |
|--------------------------------------|------------------|----------------------|-----------------------|-----------------|-------------|-----------------------------------|------------|-------------------------------|------|
| | | | >2000 μ | 1180-2000 μ | <1180 μ | 1st 3 days | 1st 7 days | Average %/Day after 7 days | Days |
| 1-9 (7-032) | 63.6 | 62 | 15.6 | 81.0 | 3.4 | 11.4 | 15.4 | 0.326 | 87 |
| 1-10A | 63.2 | 80 | 8.8 | 81.6 | 9.6 | 5.9 | 12.2 | 0.414 | 84 |
| 1-10B | 59.3 | 58 | -- | -- | -- | 11.8 | 16.7 | 0.354 | 84 |
| 1-11E (7-033/7-034) | 64.1 | 67 | 24.6 | 69.7 | 5.7 | 5.8 | 8.2 | 0.346 | 84 |
| 1-12B | 66.2 | 36 | 4.7 | 88.0 | 7.3 | 21.3 | 23.0 | 0.259 | 84 |
| 1-12C | 66.8 | 38 | 3.1 | 95.8 | 1.1 | 24.3 | 26.1 | 0.200 | 84 |
| 1-14 | 64.5 | 64 | 0.5 | 92.4 | 7.1 | 22.9 | 28.2 | 0.220 | 81 |
| 1-15A | 64.0 | 38 | 0.6 | 91.9 | 7.5 | 20.2 | 22.2 | 0.249 | 80 |
| 1-16 | 63.9 | 24 | 1.9 | 89.3 | 8.8 | 3.6 | 4.9 | 0.246 | 79 |
| 1-17 | 63.4 | 23 | 12.6 | 81.2 | 6.2 | 1.3 | 1.9 | 0.138 | 78 |
| 1-18A2 | 65.0 | 32 | 11.6 | 81.2 | 7.2 | 0.99 | 1.9 | 0.199 | 73 |
| 1-18B ** | 63.9 | 44 | 16.3 | 82.1 | 1.6 | 0.97 | 1.6 | 0.112 | 73 |
| 1-20A1 ** | 65.1 | 46 | 10.3 | 77.7 | 12.0 | 2.2 | 3.7 | 0.205 | 72 |
| 1-20A2 ** | 64.0 | 41 | 23.6 | 72.5 | 3.9 | 3.8 | 5.9 | 0.166 | 72 |
| 1-20B1 | 65.5 | 50 | 24.5 | 68.3 | 7.2 | 1.2 | 2.2 | 0.132 | 72 |
| 1-20B2 | 65.5 | 46 | 12.3 | 80.0 | 7.7 | 1.5 | 3.0 | 0.194 | 72 |
| 1-23A * | 62.2 | 18 | 66.1 | 27.2 | 6.7 | 7.4 | 8.5 | 0.119 | 65 |

(1) Indicates samples submitted to Bureau of Mines

* Runs using basic formulation

** Runs using polyethylene with paraffin and Piccolyte.

*** Runs using formulation of choice

Table II (Cont'd)

| Run No. (SwRI No.) | Payload wt. % | Burst Strength, g | Size Distribution - % | | | Water Loss Rate - % Original Fill | | | |
|--------------------------|------------------|----------------------|-----------------------|-----------------|-------------|-----------------------------------|------------|-------------------------------|------|
| | | | >2000 μ | 1180-2000 μ | <1180 μ | 1st 3 days | 1st 7 days | Average %/Day after 7 days | Days |
| 1-23B * | 62.2 | 14 | 40.1 | 51.4 | 8.5 | 4.1 | 4.9 | 0.219 | 65 |
| 1-24A | 61.0 | 39 | 53.1 | 44.6 | 2.3 | 7.4 | 9.4 | 0.265 | 64 |
| 1-27A ** | 61.2 | 28 | 16.5 | 74.5 | 9.0 | 9.1 | 10.9 | 0.235 | 59 |
| 1-27B1 | 62.5 | 43 | 8.2 | 86.6 | 5.2 | 2.1 | 3.1 | 0.170 | 59 |
| 1-27B2 | 60.1 | 52 | 12.7 | 82.1 | 5.2 | 3.2 | 4.0 | 0.182 | 59 |
| 1-27C ** | 63.1 | 31 | 0.8 | 88.6 | 10.6 | 6.5 | 9.6 | 0.249 | 59 |
| 1-28A1 ** | 61.7 | 44 | 3.1 | 85.8 | 11.1 | 6.0 | 7.5 | 0.214 | 59 |
| 1-28A2 ** | 62.3 | 40 | 4.0 | 87.6 | 8.4 | 3.5 | 5.0 | 0.208 | 59 |
| 1-28A3 ** | 62.4 | 47 | 6.5 | 86.8 | 6.7 | 2.4 | 3.5 | 0.179 | 59 |
| 1-28B1 ** | 64.8 | 25 | 8.0 | 90.0 | 2.0 | 1.4 | 2.2 | 0.160 | 59 |
| 1-28B2 ** | 65.3 | 42 | 2.8 | 91.9 | 5.3 | 2.7 | 4.7 | 0.233 | 59 |
| 1-28C | 63.1 | 21 | 8.5 | 79.9 | 11.6 | 4.5 | 4.9 | 0.058 | 59 |
| 1-32A1 ** | 64.6 | 57 | 42.1 | 56.0 | 1.9 | 1.1 | 1.7 | 0.143 | 56 |
| 1-32A2 ** | 66.4 | 60 | 18.3 | 78.0 | 3.7 | 1.4 | 2.0 | 0.154 | 56 |
| 1-32B1 | 61.4 | 35 | 9.2 | 74.8 | 16.0 | 18.1 | 21.7 | 0.346 | 17 |
| 1-32B2 | 65.3 | 33 | 2.8 | 86.5 | 10.7 | 21.2 | 24.7 | 0.203 | 56 |
| 1-35A1 | 65.6 | 50 | 7.3 | 84.8 | 7.9 | 1.8 | 3.7 | 0.170 | 51 |
| 1-35A2 | 65.8 | 40 | 11.4 | 84.1 | 4.5 | 0.9 | 2.0 | 0.153 | 51 |
| 1-36 | 65.5 | 57 | 38.4 | 60.2 | 1.4 | 0.9 | 1.7 | 0.102 | 59 |
| 1-38A * | 64.8 | 36 | 2.5 | 95.8 | 1.7 | 2.7 | 3.2 | 0.183 | 46 |
| 1-38B * (7-059/7-060) | 64.3 | 25 | 4.0 | 90.4 | 5.6 | 1.2 | 2.1 | 0.091 | 45 |
| 1-38C * | 63.9 | 21 | 29.1 | 68.5 | 2.4 | 0.9 | 1.7 | 0.060 | 45 |

Table II (Cont'd)

| Run No. (SwRI No.) | Payload wt. % | Burst Strength, g | Size Distribution - % | | | Water Loss Rate - % Original Fill | | | |
|-----------------------|------------------|----------------------|-----------------------|-----------------|-------------|-----------------------------------|------------|-------------------------------|------|
| | | | >2000 μ | 1180-2000 μ | <1180 μ | 1st 3 days | 1st 7 days | Average %/Day after 7 days | Days |
| 1-40 | 65.2 | 19 | 24.9 | 73.7 | 1.4 | 1.5 | 1.9 | 0.011 | 42 |
| 1-42B * | not tested | | 94.3 | 5.7 | nil | not tested | | | |
| 1-43A * | not tested | | 99.0 | 1.0 | nil | not tested | | | |
| 1-43B * | not tested | | 88.6 | 9.2 | 2.2 | not tested | | | |
| 1-43C | not tested | 20.5 | 70.8 | 26.6 | 2.6 | not tested | | | |
| 1-45 ** | not tested | 43 | 71.1 | 27.7 | 1.3 | not tested | | | |
| 1-46 (7-061) | 64.7 | 58 | 30.2 | 68.4 | 1.4 | 2.4 | 4.6 | | |
| | | | | | | 1.7 | 3.5 | | |
| | | | | | | 1.9 | 3.7 | | |
| | | | | | | 2.2 | 4.1 | | |
| | | | | | | 1.8 | 3.3 | | |
| | | | | | | Avg. 2.0 | 3.8 | 0.089 | 153 |
| 1-47A * | 60.2 | no test | 38.9 | 47.4 | 13.7 | 5.1 | 5.7 | 0.041 | 148 |
| 1-47B * | 61.3 | 32 | 16.2 | 73.0 | 10.8 | 1.7 | 2.8 | 0.060 | 147 |
| 1-49A * | 64.2 | no test | 77.8 | 21.0 | 1.2 | 1.4 | 2.4 | 0.021 | 144 |
| 1-49B * | 63.5 | no test | 68.2 | 29.0 | 2.8 | 1.9 | 3.0 | 0.022 | 144 |
| 1-49C * | 60.2 | no test | 55.6 | 41.5 | 2.9 | 1.8 | 2.9 | 0.040 | 144 |
| 1-50A * | no tests | | 62.0 | 35.6 | 2.4 | no tests | | | |
| 1-50B * | no tests | | 68.7 | 29.8 | 1.5 | no tests | | | |
| 1-51A * | 60.5 | 28 | 70.0 | 28.2 | 1.8 | 7.1 | 8.8 | 0.026 | 138 |
| 1-51B1 | 59.7 | 28 | 61.6 | 35.7 | 2.7 | 12.8 | 13.8 | 0.020 | 138 |
| 1-51B2 | 63.2 | 29 | 54.1 | 41.6 | 4.3 | 2.2 | 3.4 | 0.012 | 138 |
| 1-51C1 | no tests | | 80.2 | 18.9 | 0.9 | no tests | | | |
| 1-51C2 | 59.9 | 62 | 57.4 | 39.4 | 2.3 | 7.6 | 10.9 | 0.020 | 138 |
| 1-53A * | 61.4 | -- | 76.1 | 22.3 | 1.6 | 4.6 | 5.3 | 0.012 | 130 |

CS

Table II (Cont'd)

| Run No. (SwRI No.) | Payload wt. % | Burst Strength, g | Size Distribution - % | | | Water Loss Rate - % Original Fill | | | |
|-----------------------|------------------|----------------------|-----------------------|-----------------|-------------|-----------------------------------|------------|-------------------------------|------|
| | | | >2000 μ | 1180-2000 μ | <1180 μ | 1st 3 days | 1st 7 days | Average %/Day after 7 days | Days |
| 1-53B * | 59.6 | -- | 80.2 | 18.8 | 1.0 | 5.2 | 6.2 | 0.004 | 130 |
| 1-53C * | 60.9 | -- | 84.6 | 14.6 | 0.8 | 3.6 | 4.2 | 0.001 | 130 |
| 1-53D * | 61.4 | -- | 85.0 | 14.4 | 0.6 | no tests | | | |
| 1-55A | no tests | | | | | | | | |
| 1-55B | 63.8 | 32 | 65.5 | 32.9 | 1.6 | 4.4 | 4.6 | 0.013 | 127 |
| 1-55C | 64.1 | 37 | 74.9 | 23.9 | 1.2 | 7.8 | 9.3 | 0.036 | 127 |
| 1-56A *** | 63.3 | 24 | 54.8 | 43.1 | 2.1 | 1.1 | 1.4 | 0.016 | 125 |
| 1-56B *** | 61.7 | 33 | 31.5 | 60.9 | 7.6 | 0.4 | 0.6 | 0.012 | 125 |
| 1-56C | 61.1 | 76 | 50.8 | 47.1 | 2.1 | 1.6 | 1.8 | 0.018 | 125 |
| 2-2 | 61.9 | 65 | 47.6 | 50.1 | 2.3 | 0.5 | 1.8 | 0.012 | 123 |
| 2-4A *** | 59.4 | -- | 83.1 | 16.6 | 0.3 | 0.2 | 0.4 | 0.006 | 181 |
| 2-4B *** | 61.4 | -- | 67.1 | 31.0 | 1.8 | 0.2 | 0.3 | 0.007 | 181 |
| 2-4C *** | 61.9 | 26 | 71.2 | 28.7 | 0.1 | 0.2 | 0.3 | 0.011 | 181 |
| 2-6A *** | 61.2 | -- | 75.7 | 22.8 | 1.4 | 3.6 | 4.7 | 0.025 | 181 |
| 2-6B *** | 62.2 | -- | 78.8 | 20.1 | 1.1 | 0.5 | 0.7 | 0.006 | 181 |
| 2-8 *** (7-079) | 62.5 | -- | 21.6 | 77.2 | 1.2 | 1.3 | 1.8 | 0.029 | 159 |
| 2-9A | 66.2 | ca.19 | 13.8 | 82.8 | 3.4 | 0.6 | 1.0 | 0.030 | 158 |
| 2-9B1 | 65.0 | 20 | 17.0 | 82.2 | 0.8 | 1.2 | 1.6 | 0.009 | 158 |
| 2-9B3 | 61.2 | 18 | 34.3 | 64.8 | 0.9 | 0.3 | 0.5 | 0.008 | 158 |
| 2-12A | 67.1 | 22 | 6.3 | 92.4 | 1.3 | 1.6 | 2.0 | 0.059 | 153 |
| 2-12B ** | 61.4 | 62 | 27.0 | 71.9 | 1.1 | 3.1 | 5.0 | 0.059 | 153 |

Table II (Cont'd)

| Run No. (SwRI No.) | Payload wt. % | Burst Strength, g | Size Distribution - % | | | Water Loss Rate - % Original Fill | | | |
|-----------------------|------------------|----------------------|-----------------------|-----------------|-------------|-----------------------------------|------------|-------------------------------|------|
| | | | >2000 μ | 1180-2000 μ | <1180 μ | 1st 3 days | 1st 7 days | Average %/Day after 7 days | Days |
| 2-14A ** | 63.3 | 23 | 19.4 | 80.4 | 0.2 | 1.3 | 1.8 | 0.063 | 152 |
| 2-14B | 59.9 | 26 | 23.6 | 76.0 | 0.4 | 8.6 | 9.3 | 0.051 | 152 |
| 2-20A | 63.6 | | 27.8 | 71.1 | 1.1 | 1.0 | 1.2 | 0.001(gain) | 133 |
| 2-20B | 65.0 | 28 | 18.3 | 79.8 | 1.9 | 0.8 | 1.0 | 0.001(gain) | 133 |
| 2-21 | 64.7 | | 0.4 | 91.2 | 8.4 | 1.5 | 1.7 | 0.020 | 132 |
| 2-22A | 69.3 | 14 | 20.5 | 76.8 | 2.7 | 0.5 | 0.7 | 0.015 | 131 |
| 2-22C * | 69.3 | 16 | 47.0 | 51.2 | 1.8 | 0.8 | 1.0 | 0.021 | 131 |
| 2-24A * | 68.0 | | 44.4 | 53.2 | 2.4 | 2.6 | 3.0 | 0.035 | 129 |
| 2-24B *** | 68.5 | 10 | 16.7 | 82.4 | 0.9 | 0.4 | 0.5 | 0.026 | 129 |
| 2-24C *** | 64.8 | 13 | 10.3 | 88.9 | 0.8 | 0.3 | 0.4 | 0.020 | 129 |
| 2-26 | 66.8 | 22 | 20.6 | 76.6 | 2.8 | 0.8 | 1.0 | 0.018 | 125 |
| 2-29A *** | 65.8 | -- | 18.3 | 80.2 | 1.5 | 0.5 | 0.6 | 0.015 | 102 |
| 2-29B *** | 67.3 | -- | 26.7 | 71.6 | 1.7 | 1.1 | 1.6 | 0.020 | 102 |
| 2-31A *** | 63.6 | -- | 25.8 | 73.1 | 1.1 | 1.0 | 1.4 | 0.017 | 96 |
| 2-31B *** | 66.5 | -- | -- | -- | --- | 0.8 | 1.0 | 0.009 | 96 |
| 2-32 *** | 71.2 | -- | 40.8 | 58.2 | 1.0 | 2.9 | 3.2 | 0.016 | 95 |
| 2-35 *** | 68.8 | -- | 32.7 | 67.1 | 0.2 | 2.3 | 2.7 | 0.023 | 69 |
| 2-36A *** | 69.8 | -- | 47.4 | 51.3 | 1.3 | 1.1 | 1.4 | 0.022 | 68 |
| 2-36B *** | 69.1 | -- | 41.4 | 58.1 | 0.5 | 1.1 | 1.3 | 0.030 | 68 |
| 2-37A *** | 67.1 | -- | 37.1 | 62.2 | 0.7 | 0.3 | 0.5 | 0.022 | 66 |
| 2-37B *** | 69.4 | -- | 34.9 | 64.0 | 1.1 | 0.3 | 0.4 | 0.015 | 66 |
| 2-38 *** | 63.5 | 22.5 | 35.2 | 64.5 | 0.3 | 0.1 | 0.3 | 0.015 | 66 |

Table II (Cont'd)

| Run No. (SwRI No.) | Payload wt. % | Burst Strength, g | Size Distribution - % | | | Water Loss Rate - % Original Fill | | | |
|-----------------------|------------------|----------------------|-----------------------|-----------------|-------------|-----------------------------------|------------|-------------------------------|------|
| | | | >2000 μ | 1180-2000 μ | <1180 μ | 1st 3 days | 1st 7 days | Average %/Day after 7 days | Days |
| 2-39A *** | 64.8 | -- | 47.3 | 52.1 | 0.6 | 0.4 | 0.5 | 0.008 | 62 |
| 2-39B *** | 64.4 | -- | 41.2 | 58.2 | 0.6 | 0.6 | 0.7 | 0.007 | 62 |
| 2-41A *** | 66.2 | -- | 45.7 | 53.6 | 0.7 | 0.6 | 0.8 | 0.010 | 62 |
| 2-41B *** | 66.2 | -- | 37.2 | 59.6 | 3.2 | 0.5 | 0.6 | 0.010 | 62 |
| 2-42A *** | 67.6 | | 65.0 | 32.8 | 2.2 | 3.4 | 3.8 | 0.008 | 59 |
| 2-42B *** | 69.4 | | 73.2 | 25.9 | 0.9 | 2.8 | 3.3 | 0.013 | 59 |
| 2-44A *** | 65.6 | | 35.1 | 64.1 | 0.8 | 0.4 | 0.6 | 0.023 | 31 |
| 2-44B *** | 65.1 | | 18.3 | 78.4 | 3.3 | 0.8 | 1.1 | 0.035 | 31 |
| 2-45 *** | 63.8 | | 11.1 | 75.5 | 13.4 | 2.4 | 2.8 | 0.041 | 31 |
| 2-46 | 58.8 | | 18.2 | 74.3 | 7.5 | 1.1 | 1.5 | 0.046 | 27 |
| 2-47A | 58.7 | | 24.5 | 65.5 | 10.0 | 6.4 | 7.0 | 0.059 | 27 |
| 2-47B | 57.7 | | 26.5 | 61.2 | 12.3 | 3.3 | 3.7 | 0.062 | 27 |
| 2-47C | 57.1 | | 21.8 | 64.8 | 13.4 | 4.2 | 4.6 | 0.038 | 27 |
| 2-49A | 55.6 | -- | 14.9 | 71.0 | 14.1 | no test | | | |
| 2-49B | -- | -- | 42.7 | 48.7 | 8.6 | no test | | | |
| 2-50A | 59.4 | -- | 72.0 | 26.9 | 1.1 | no test | | | |
| 2-50B | 57.9 | -- | 57.3 | 39.2 | 3.5 | no test | | | |
| 2-50C | 61.1 | -- | 63.4 | 33.7 | 2.9 | no test | | | |
| 2-51A | 62.1 | -- | 27.3 | 65.6 | 7.1 | 3.4 | 4.1 | 0.077 | 13 |
| 2-51B | 65.9 | -- | 41.6 | 52.3 | 6.1 | 4.0 | 4.7 | 0.057 | 13 |
| 2-52 | 65.9 | -- | 26.5 | 65.1 | 8.4 | 4.9 | 5.9 | 0.072 | 12 |

Table II (Cont'd)

| Run No. (SwRI No.) | Payload wt. % | Burst Strength, g | Size Distribution - % | | | Water Loss Rate - % Original Fill | | | |
|-----------------------|------------------|----------------------|-----------------------|-----------------|-------------|-----------------------------------|------------|-------------------------------|------|
| | | | >2000 μ | 1180-2000 μ | <1180 μ | 1st 3 days | 1st 7 days | Average %/Day after 7 days | Days |
| 2-53A | 61.5 | -- | 31.6 | 60.3 | 8.1 | 4.7 | 5.7 | 0.085 | 11 |
| 2-53B | 61.9 | -- | 23.5 | 65.8 | 10.7 | 3.1 | 4.0 | 0.094 | 11 |
| 2-53C | 60.4 | -- | 32.8 | 60.3 | 6.9 | 4.2 | 5.4 | 0.080 | 11 |
| 2-53D | 60.7 | -- | 18.1 | 71.2 | 10.7 | 4.5 | 6.0 | 0.124 | 11 |
| 2-53E | 58.5 | -- | 26.5 | 65.6 | 7.9 | 3.4 | 4.5 | 0.081 | 11 |
| 3-1A | 62.5 | -- | 13.3 | 75.6 | 11.1 | 6.4 | 8.5 | 0.214 | 7 |
| 3-1B | 61.1 | -- | 22.1 | 67.6 | 10.3 | 4.0 | 5.3 | 0.214 | 7 |
| 3-3 (7-085) | 61.3 | -- | -- | -- | -- | 4.4 | 4.7 | 0.041 | 6 |
| 3-4A | 64.8 | -- | 10.4 | 81.8 | 7.8 | 1.5 | 3.0 | 0.131 | 4 |
| 3-4B | 65.7 | -- | 34.9 | 64.0 | 1.1 | 1.4 | 1.9 | 0.068 | 4 |
| 3-5 | 64.9 | -- | 18.7 | 76.0 | 5.3 | 1.0 | 1.6 | 0.072 | 4 |
| 3-6A | 60.7 | -- | 22.9 | 75.5 | 1.6 | 2.7 | 3.2 | 0.117 | 3 |
| 3-6B | 64.5 | -- | 17.1 | 76.8 | 6.1 | 2.3 | 2.8 | 0.094 | 3 |

APPENDIX B
Blocking Tests

A. Room Temperature Tests - ca. 75 F

1. Capsules from Run 2-14A* were placed in a glass pipe, 1-7/16 inches inside diameter, to a height of 34 inches. No water was added. Pipe was stoppered and held vertical for 36 days. Stoppers were removed and capsules were dislodged by tapping on the pipe. Only slight blocking was apparent, and capsules probably would have poured out easily from a larger diameter pipe.
2. Capsules from Run 2-8* were placed in a PVC pipe two inches ID to a height of 34 inches (1072 grams of capsules). Held 23 days. Emptied. Completely free-flowing with slight tapping.
3. Used capsules from Run 2-21* as in (2) above. Held 22 days in the Only a little harder to free than Run 2-8 capsules. Free-flowing.
4. Used capsules from Run 2-22C* and Run 2-24A* mixed as in (2) above. Held 20 days in the same manner as Run 2-8. Freed about as easily as 2-21. Free-flowing.

B. Elevated Temperature Tests

1. About 15 grams of capsules in vials were held in water bath for about 30 minutes at varied temperatures and observed upon removal and on cooling to give the following results.

| <u>Run No.*</u> | <u>Little or No Blocking, °C</u> | <u>Blocks But Parts, °C</u> | <u>Blocks and Parts with Obvious Damage, °C</u> |
|-----------------|----------------------------------|-----------------------------|---|
| 1-53A | No determination | 43-46 | 47 |
| 2-8 | 48 | 48-53 | 56 |
| 2-9A | No determination | 47-52 | 56 |
| 2-9B3 | No determination | 43-53 | 56 |
| 2-12A | 46 | 47-52 | 56 |
| 2-12B | 49 | 52-60 | No determination |
| 2-14A | 49 | 52 | 56 |
| 2-14B | 49 | 52 | 56 |

2. Samples of capsules from two runs were held at 53 to 54°C for 30 minutes. After cooling, the capsules were parted with no obvious damage and gave following results by the weight loss tests.

Run 2-8 3.9% loss of original fill first 3 days
 4.1% loss of original fill first 7 days
 Average loss 0.016%/day for 17 days after first 7 days.

Run 2-9B3 20.0% loss of original fill first 3 days
 20.6% loss of original fill first 7 days
 Average loss 0.037%/day for 17 days after first 7 days.

C. Agents to Inhibit Blocking

1. Procedure

10 grams of capsules from 2-9B3 were mixed with a small amount of candidate powder and held in a vial at 54 C for 30 minutes.

2. Results

- a. Microthene 500 - micronized polyethylene - blocked same as control
- b. Dry Flo Starch - hydrophobic starch - blocked same as control.
- c. Vulca 90 - water insoluble starch - blocked same as control.
- d. Silanox - hydrophobic micronized silica - blocked same as control.
- e. Cab-O-Sil - micronized silica - still hard to part but not as bad as control.

* Shell composition of capsules used.

| | | | | | |
|-------|-----------------|-----|-------|-----------------|-----|
| 1-53A | Sunoco 4412 | 40% | 2-12B | Boler 1426 | 50% |
| | Piccolyte S-115 | 47 | | Piccolyte S-115 | 45 |
| | Polybutene f28 | 13 | | Epolene C-16 | 5 |
| 2-8 | Sunoco 5512 | 45 | 2-14A | Sunoco 4412 | 30 |
| | Piccolyte S-115 | 45 | | Boler 1426 | 20 |
| | Polybutene 128 | 10 | | Piccolyte S-115 | 45 |
| | | | | Epolene C-16 | 5 |
| 2-9A | Boler 1435 | 45 | | | |
| | Piccolyte S-115 | 45 | 2-14B | Sunoco 4412 | 25 |
| | Polybutene 128 | 10 | | Boler 1426 | 20 |
| | | | | Piccolyte S-115 | 45 |
| 2-9B3 | Boler 1426 | 45 | | Epolene C-16 | 5 |
| | Piccolyte S-115 | 45 | | Polybutene 128 | 5 |
| | Polybutene 128 | 10 | | | |
| | | | 2-21 | Same as 2-9B3 | |
| 2-12A | Boler 1426 | 53 | | | |
| | Piccolyte S-115 | 47 | 2-22C | Same as 1-53A | |
| | | | 2-24A | Same as 1-53A | |