

## Composites Obtained by Encapsulation and Collimation of Glass Fibers Within a Thermoplastic Matrix by Means of Polymerization

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### Synopsis

Styrene and acrylonitrile monomers are polymerized in an aqueous suspension system in the presence of chopped glass strands. Under certain conditions, in the course of SAN polymerization, it is possible to align the glass strands in a parallel array (collimate) to form capsules varying in shape from tapes to cylinders. The parallel stacking of glass strands within the capsules allows for high glass loadings and results in glass microfibers completely wetted and imbedded in the polymeric matrix. The mechanism of encapsulation and collimation is discussed and is shown to be critically affected by several factors, the most important being: the preferential wetting of the strands by the monomers, the nature of the glass surface, the monomer composition, the type and concentration of protective colloid used in the suspension polymerization, and the type and intensity of agitation. The fiber glass composites obtained by encapsulation are found to have mechanical properties superior to those obtainable by conventional commercial routes.

### INTRODUCTION

Short discontinuous glass fibers are extensively used to upgrade the mechanical properties of thermoplastics.

Fiber glass-reinforced composites of poly(styrene-co-acrylonitrile), abbreviated SAN, are commercially prepared by two general routes<sup>1-4</sup>: (A) dry blending of chopped glass strands with an SAN matrix, and (B) by blending SAN with a SAN glass concentrate produced by coating and impregnation of continuous glass rovings, followed by fusion of the polymer and chopping of the resulting impregnated strands.

Route (A) is considered more economical in large-volume operations. However, it requires high capital investment for the equipment necessary for feeding and proportionating the feed of glass strands and matrix in order to avoid severe problems of nonuniformity of glass distribution, and of glass segregation, bridging, and "haystacking" during blending, feeding, and processing.

Route (B) is considered more economical for lower-volume operations because of lower capital investment requirements, despite the considerably higher cost of the pellets. Route (B) results in pellets of convenient size for molding and blending, where the coated strands are oriented parallel to each other in the longitudinal direction. Such parallel stacking of glass fibers allows for economi-

cally desirable high glass loadings (glass concentrates) which cannot be produced by the random "haystacking" of glass strands by the dry blending route (A). Route (B), however, may lead to an uneven penetration of the glass roving by the viscous polymer. This results in a concentration gradient of SAN decreasing in the direction of the glass roving core which may remain incompletely wetted and impregnated by the polymer.

Our work<sup>6-9</sup> had the following objectives: (1) to investigate economical methods for obtaining high loadings of glass fibers in SAN pellets of desirable size, (2) to achieve thorough wetting of glass fibers by the matrix and uniform distribution of matrix within the pellets, and (3) to improve composite properties.

The present publication confines itself with encapsulation and collimation (i.e., alignment of fibers in a parallel array) of discontinuous glass strands in SAN.

Encapsulation and collimation of boron or glass fibers in thermosetting epoxy resins was previously described by Andersen and Morris.<sup>5</sup> This was achieved by coalescence and deposition of an emulsion or suspension epoxy resin on the surface of water-wetted fibers.

## EXPERIMENTAL

**Encapsulation of Glass Strands.** Encapsulation of  $1/8$ - or  $1/4$ -in.-long chopped glass strands was conducted by polymerizing S/AN monomers in the presence of the loose strands in 28-32 oz pressure bottles.

**Charge.** The following is a typical formulation for the encapsulation of 50%  $1/8$ -in. glass strands in 74S/26AN:

Ingredients	Charge, g	Parts/100 parts of (monomers + glass)
$1/8$ " Strands <sup>a</sup>	80.00	50.0
Styrene	59.20	37.0
Acrylonitrile	20.80	13.0
Benzoyl peroxide	0.40	
Terpinolene	0.24	
Protective colloid A <sup>b</sup>	0.48	0.3
or		
Protective colloid B <sup>c</sup>	1.60	1.0
Distilled water	420.00	262.0

<sup>a</sup> Johns-Manville's CS-308.

<sup>b</sup> Colloid A = copolymer of 95.5 mole-% acrylic acid and 4.5 mole-% 2-ethylhexyl acrylate.

<sup>c</sup> Colloid B = Bentonite KWK (American Colloid Co.). The pH of the aqueous solution was adjusted to 6.0 by addition of 0.5*N* sulfuric acid.

**Procedure.** The reactors are charged with glass strands and the air is removed by alternating application of vacuum with breaking of the vacuum with nitrogen. Nitrogen-purged solutions of initiator and chain transfer agent in the S/AN monomers are introduced under nitrogen. The reactors are sealed for a short time to permit thorough wetting of the strands by the monomers, and a nitrogen-purged aqueous solution of colloid A or B is then introduced under nitrogen. The reactors are sealed and polymerization is conducted at 75°C by rotating the reactor end-over-end at 33 rpm in a water bath. After a cycle of 15-17 hr, the contents are discharged on a filter, washed repeatedly with

water, leached in hot methanol (to remove any unreacted monomers), and dried at 75°C in vacuo. Essentially complete conversion is obtained.

The % glass in the capsules was determined by burning off of the matrix from the capsules in a muffle furnace.

The desired 20% by weight of glass in the composite was achieved by appropriate dilution with a preformed 74S/26AN copolymer of 0.83 intrinsic viscosity in DMF at 30°C. The intrinsic viscosity of the SAN matrix formed during encapsulation, controlled by use of terpinolene as the chain transfer agent, was in the same range of the SAN diluent.

**Processing of Composites.** The blends of glass strands or encapsulated glass and SAN diluent, adjusted to give 20% by weight of glass, were extruded at 425–450°F in a 1-in. extruder, using a single-flight vinyl screw having an  $L/D$  ratio of 18, and pelletized. The resulting pellets were injection molded at 450°F by using a nozzle with oversize orifice and a die with wide gates.

**Testing of Composites.** Tensile properties were obtained on 4 in.  $\times$   $1/4$  in.  $\times$   $1/10$  in. test specimens. Tensile modulus and fail elongation were determined by using an extensometer.

Izod impact strength was obtained on  $2\frac{1}{2}$  in.  $\times$   $1/2$  in.  $\times$   $1/8$  in. specimens notched with 0.010 in. notch radius.

Heat distortion temperature was obtained on  $2\frac{1}{2}$  in.  $\times$   $1/2$  in.  $\times$   $1/8$  in. specimens, at 264 psi fiber stress, with a 2-in. span.

## DISCUSSION

### Encapsulation and Collimation of Discontinuous Glass Strands in SAN

The encapsulation process involves polymerizing styrene and acrylonitrile in the presence of loose,  $1/8$ -in.- to  $1/4$ -in.-long strands in an aqueous suspension system. Capsules are obtained consisting of glass strands completely imbedded and "cemented" in the polymerized matrix. In the course of polymerization, the strands align themselves in a parallel array (collimate) to form capsules which can vary in shape from flat tapes to perfect cylinder or skewed cylinders. The glass fibers are generally butted end-to-end, the length of the capsules being essentially the same, or slightly greater, than the original strand. The microfibers of the strands (the strands being composed of about 200–300 microfibers, about 0.00035 in. in diameter, arranged in parallel configuration) are fully wetted and coated by the imbedding polymeric matrix. Under certain conditions, capsules of ellipsoidal shape are obtained where the fibers are still arranged in a parallel array.

The encapsulation process is critically affected by several factors, the most important being: the order of wetting the glass strands; the type and concentration of protective colloid used during polymerization; the nature of the sizing agent used to prepare and protect the original glass strands; the length of the strands; the ratios of monomer to water and glass to monomer; monomer composition; and type and intensity of agitation.

Some of the most critical factors affecting encapsulation and collimation will be discussed in some detail.

### Type and Concentration of Protective Colloid

The composition and concentration of protective colloid (suspending agent) critically affects the encapsulation process. Depending on conditions used, a

TABLE I  
Effect of Protective Colloid Concentration on Glass Strand  
Encapsulation in SAN (74S/26AN)

Glass strand size, in. <sup>a</sup>	Mon./glass wt ratio	% Colloid based on (mon. + glass)	Size of capsules <sup>b</sup>	Product appearance	
1/8	4/1	0.052 colloid A	1-2.5 cm	not collimated	oblong beads
1/8	4/1	0.078	20-100X	collimated	
1/8	4/1	0.104	10-20X	collimated	
1/8	4/1	0.20	4-10X	collimated	many fines <sup>c</sup>
1/8	1/1	0.25	80-150X	collimated	few fines
1/8	1/1	0.35	40-120X	collimated	few fines
1/8	1/1	0.40	5-100X	collimated	few fines
1/8	1/1	0.50	4-20X	collimated	many fines
1/4	4/1	0.125	large flat oval beads	—	—
1/4	4/1	0.175	large flat oval beads	—	—
1/4	4/1	0.20	100-300X	collimated	few fines
1/4	4/1	0.25	100-300X	collimated	few fines
1/8	4/1	0.20 colloid B		agglomeration	—
1/8	4/1	0.30	10-40X	collimated	few fines
1/8	4/1	0.40	5-20X	collimated	many fines
1/8	4/1	0.50	2-10X	collimated	many fines
1/8	1/1	0.70	agglomerated		
1/8	1/1	0.85	40-100X		
1/8	1/1	1.0	40-100X		
1/8	1/1	none	agglomeration		

<sup>a</sup> CS-308 (Johns Manville).

<sup>b</sup> X = Approximate number of times of increase in weight of capsule, compared to original single strand weight.

<sup>c</sup> Fines = Small SAN suspension beads devoid of glass.

range of encapsulated structures have been obtained, the two extreme cases being (1) complete agglomeration of the glass/SAN mass into large lumps, and (2) formation of many small SAN beads with essentially complete exclusion of glass strands, the strands preserving their original size and shape while remaining practically uncoated by polymer.

In between these extremes, one can obtain collimated cylindrical capsules of the desired size. The capsules of the desired size have a weight which is about 40 to 200 times that of the original single strand. In Tables I and II, under the "capsule size" heading, the above range would be expressed as "40-200X" and would not relate to the glass content of the capsule.

A small amount of fine suspension SAN beads ("fines") which do not contain any glass are often formed concurrently, particularly at low glass loadings (i.e., at low glass/monomer ratios).

In the absence of any protective colloid, one obtains total agglomeration of glass and polymer into a large lump.

A partial ester of poly(acrylic acid) (colloid A) and a water-dispersible bentonite (colloid B) were found to be the most effective protective colloids, while generally poor encapsulation was observed with colloidal alumina, or silica or silicic acid, and with partially acetylated poly(vinyl alcohol) polymers.

TABLE II  
Effect of Protective Colloid Concentration on Glass Strand  
Encapsulation in S/AN/MAA<sup>a</sup>

Glass strand size, in. <sup>b</sup>	Mon./glass wt ratio	% Colloid A based on (mon. + glass)	% glass in capsules	Size of capsules <sup>c</sup>	Product appearance	
1/8	1/1	0.10	55	10-200X	collimated	no fines <sup>d</sup>
1/8	1/1	0.125	59	15-150X	collimated	no fines
1/8	1/1	0.150		10-150X	collimated	some fines
1/8	1/1	0.20		5-100X	collimated	some fines
1/8	1/1	0.30		5-50X	collimated	some emulsion
1/8	1/1	0.40		5-15X	collimated	some emulsion
1/8	1/2	0.075	69	1-3 cm diam.	collimated	flat capsules
1/8	1/2	0.10		0.5-2 cm diam.	collimated	flat capsules
1/8	1/2	0.125	69	5-100X	collimated	no fines
1/8	1/2	0.20		4-15X	collimated	some fines
1/8	1/2	0.25	76	4-10X	collimated	many fines
1/8	1/4	0.10	81	5-200X	collimated	no fines
1/8	1/4	0.20		5-10X	collimated	some fines
1/4	1/1	0.175		20-250X	collimated	flat tapes
1/4	1/1	0.225		20-200X	collimated	flat tapes

<sup>a</sup> Terpolymer containing 72styrene/26acrylonitrile/2methacrylic acid.

<sup>b</sup> CS-308 (Johns Manville).

<sup>c</sup> X = Approximate number of times of increase in weight of capsule, compared to original single strand weight.

<sup>d</sup> Fines = Small SAN suspension beads devoid of glass.

From Table I and II, one can readily see the critical effect of suspending agent concentration on the size and degree of collimation of the resulting capsules. Capsules have been obtained containing from 20% to 80% glass by weight.

It should be noted that all experiments given in the tables were conducted by wetting the strands with monomers before addition of the aqueous solution of the suspending agent.

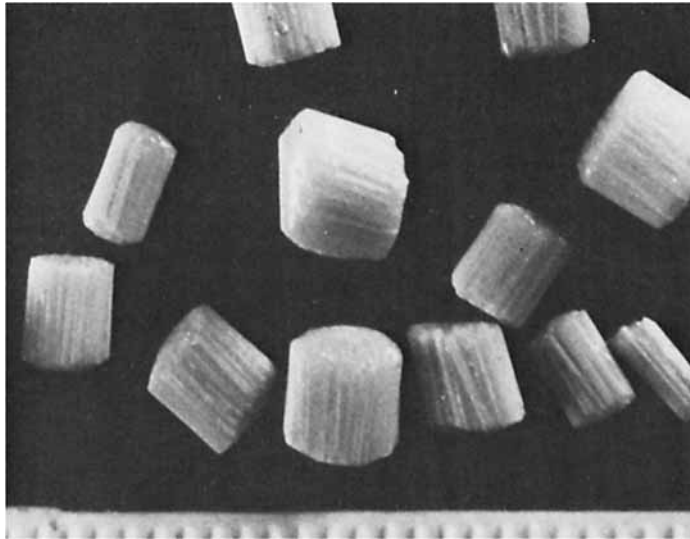
From our study of effect of protective colloid concentration, the following conclusions can be drawn:

a. For each suspending agent, there is an optimum concentration for efficient collimation and particle size control. Considerably larger amounts of colloid B than of colloid A are needed, to obtain capsule of equal size.

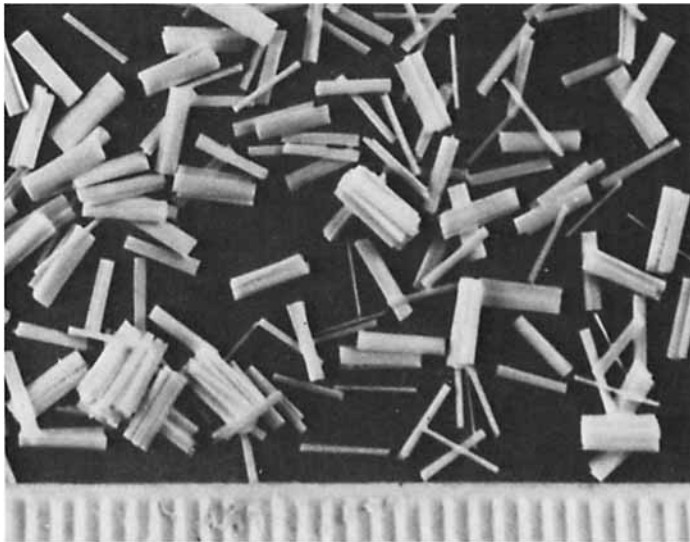
b. The optimum concentration of suspending agent is greatly dependent on the length of the strands, the ratio of monomer to glass and glass content in the formulation, and the composition of the monomers. Larger concentrations of suspending agents are required with the longer strands and with decreasing monomer/glass ratios.

c. The presence of a small amount of methacrylic acid in S/AN monomers allows considerable reduction in colloid A concentration with resulting reduced formation of SAN fines (i.e., fine SAN suspension devoid of glass).

Figures 1 and 2 are typical pictures of the collimated capsules of varying geometry obtained by varying the concentration of protective colloid. The collimated structure of the capsules can best be seen in Figure 3a, which shows a capsule coated with copper by vacuum deposition, and in Figure 3b, which shows



(a)

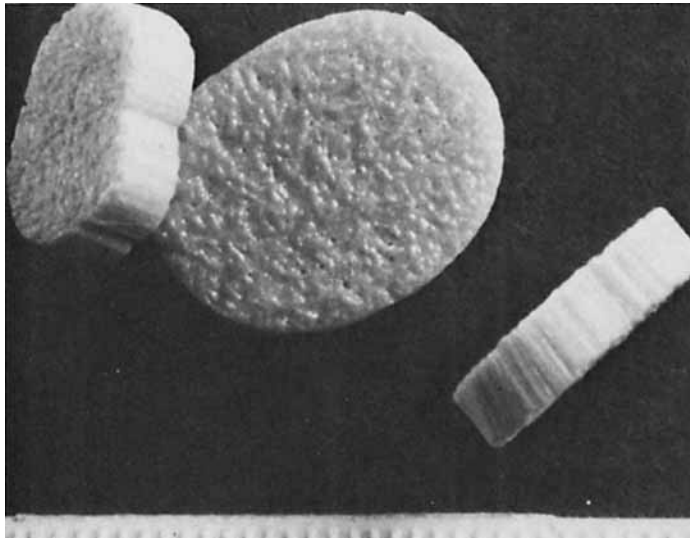


(b)

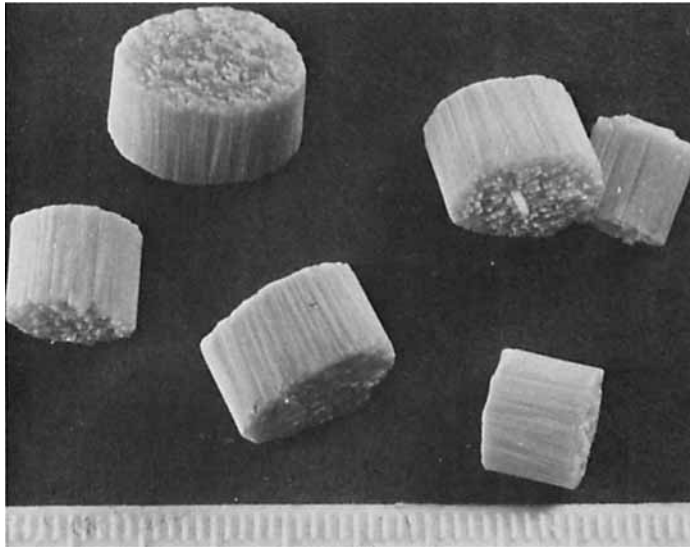
Fig. 1. Encapsulation and collimation of glass strands in SAN (smallest subdivision on ruler in pictures = 1.0 mm): (a) 60% wt of  $\frac{1}{8}$ -in. strands in SAN; (b) 71% wt of  $\frac{1}{8}$ -in. strands in SAN (excessively high concentration of colloid A).

the preservation of the collimated structure after burning away of the SAN matrix in a muffle furnace.

A few experiments conducted with styrene as the only monomer at 20% weight loadings of  $\frac{1}{8}$ -in. glass strands resulted in long, flat ellipsoidal beads with imperfect collimation. The type and concentration of suspending agent used did not appear to improve collimation. Higher glass loadings were not investigated but could lead to improved results.



(a)

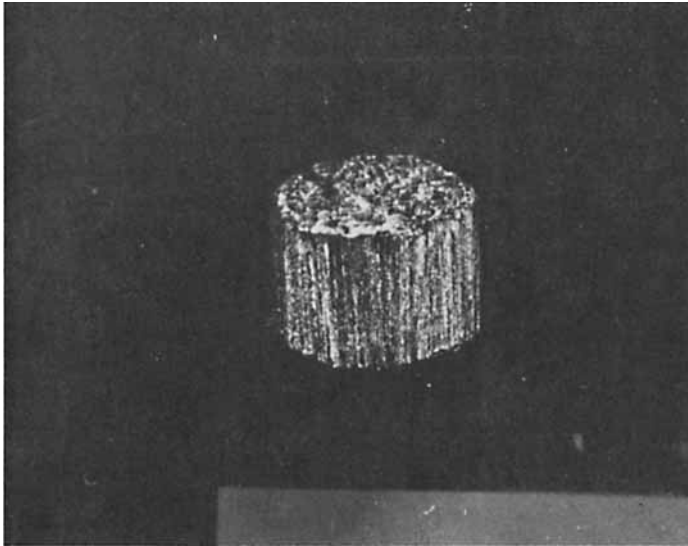


(b)

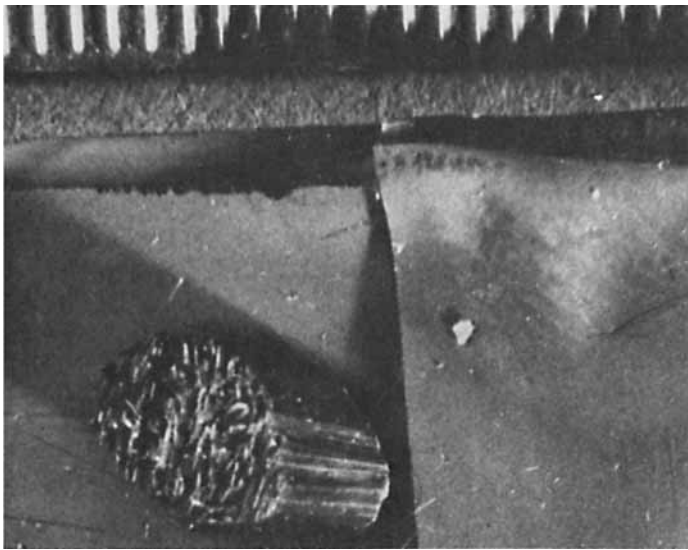
Fig. 2. Encapsulation and collimation of glass strands in SAN or S/AN/MAA (smallest subdivision on ruler in pictures = 1.0 mm): (a) 66% wt of  $1/8$ -in. strands in S/AN/MAA (excessively low colloid A concentration); (b) 71% wt of  $1/4$ -in. strands in SAN.

#### Requirements for Encapsulation and Probable Mechanism for Collimation

Wetting of the glass strands by S/AN monomers prior to wetting by water and protective colloid was found to be a necessary requirement for collimation and encapsulation in SAN. If the above order of prewetting was reversed, encapsulation of the glass strands was not achieved. Instead fine SAN suspension beads completely devoid of glass were obtained with essentially no coating of the glass.



(a)



(b)

Fig. 3. Collimated structure of capsules: (a) glass strands encapsulated in SAN (Cu coated by vacuum deposition); (b) collimated glass fiber structure preserved after burning away of SAN (Cu coated by vacuum deposition).

The sizing agent used in the manufacture of the glass strands, was found to play an important role in encapsulation. Most of the commercially available glass strands, sized with poly(vinyl acetate), were found to result in encapsulation, whereas other glass strands coated with other unidentified sizing agents generally resulted in incomplete encapsulation and in irregular capsules, quite large and oblong in shape with imperfect collimation. These results appear to be related to the relative ability of S/AN monomers to wet the surface of the glass strands. It is not clear, however, if the unsatisfactory collimation obtained



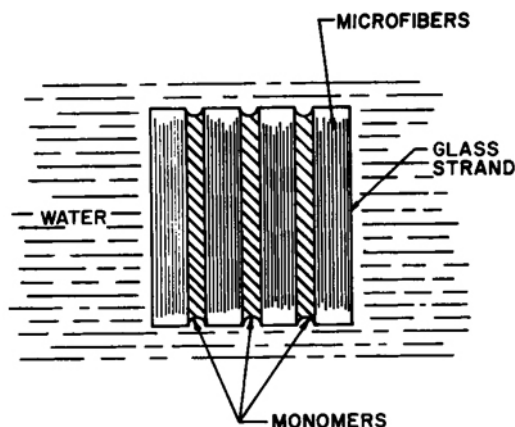


Fig. 4. Schematic representation of collimation of glass strands, prewetted by monomer, and suspended in an aqueous medium.

with other sized strands is the result of poor wetting of the strands by S/AN monomers or to the incompatibility of the glass size with the SAN forming during encapsulation.

Best results are obtained when the sizing agent becomes swollen in the monomers used to encapsulate without dissolving in the monomers.

A few experiments conducted with commercial poly(vinyl acetate) (PVA) established that S/AN monomers dissolve PVA and greatly swell the crosslinked PVA present on the surface of the glass. Microscopic examination also reveals that strands sized with PVA are more readily wetted by S/AN monomers than by water.

The above observations and results suggest that collimation is initiated by absorption of monomer by the glass strands through capillary action. Surface energy forces pull together the strands and prevent their separation. Stacks of strands thus form, possibly separated by layers of liquid monomers, which may be represented schematically as shown in Figure 4. The glass strands shown in Figure 4 contain a large number of glass microfibrils which, like the glass strands, will also imbibe monomers by capillary action.

Because of the preferential wetting by monomers, water does not displace the absorbed monomers from the glass strands during polymerization.

In addition to surface tension forces, hydrodynamic forces must come into play for the formation of collimated structures by means of agitation during the capsule growth. Gentle movement of the slurry is needed to encourage longitudinal motion and sliding of the stacks of strands to obtain collimated structures.

The mechanism of capsule growth appears similar to the behavior of logs in eddy currents of a river, where logs tend to align themselves in a parallel array to minimize the resistance to flow and are held together by hydrodynamic forces.

Collimation and capsule growth leads to a reduction in total interfacial area and is, therefore, encouraged by limited coverage by the protective colloid, whose concentration is found to be critical. Too little or no protective colloid results in total agglomeration because of insufficient protection of the surface of the growing capsule and monomer droplets. Excessive concentration of protective colloid discourages collimation because of full protection and coverage of the surface of the strands and of the suspension polymer droplets. Control of

capsule size and collimation is achieved by using a concentration of protective colloid just sufficient to protect the surface of the growing capsule.

Encapsulation was only successfully achieved with the type of low shear agitation possible with reactors gently tumbling end-over-end or with horizontal, blender-type reactors.

Excessive shear causes partial debundling of the glass strands into flexible fibrils which are unable to collimate and form large matted glass fiber balls by an "hay stacking" mechanism. Attempts to encapsulate in Pfudler-type reactors resulted in considerable glass strand debundling despite variations in agitator and baffle design aimed at minimizing localized shear intensity.

### Properties of Glass Fiber Composites Obtained by Encapsulation of Glass Strands in SAN

Representative mechanical properties of glass fiber composites obtained by encapsulation are given in Tables III and IV. The properties of dry blends of glass strands and SAN and of a commercial blend are also tabulated for purpose of comparison, all composites being compared at equal 20 weight-% glass loading. The properties of the SAN control (in the absence of glass) are also included.

The properties of dry blend composites given in Table III are representative of the properties obtained by processing of blends of chopped glass strands and an SAN matrix in the form of a powder, or crushed suspension beads, or extruded chips. In our work, direct injection molding of the above blends did not result in the indicated<sup>4</sup> property advantage over blends obtained by extrusion, followed by injection molding.

Composites prepared by encapsulation have significantly superior tensile properties and modulus and are also found (Table V) to be relatively insensitive to the processing method and to intensity of mechanical working. However, some loss in tensile strength and modulus is experienced when, following extrusion, compression molding is used in place of injection molding. This loss can

TABLE III  
Representative Properties of Glass/SAN Composites  
(All Composites were Extruded and Injection Molded)

Identification	% Glass	Tensile at fail, psi $\times 10^{-3}$	% Elongation at fail	Elastic modulus, psi $\times 10^{-5}$	Heat distortion tempera- ture at 264 psi, °C	Izod impact, ft-lb/in.
SAN copolymer <sup>a</sup>	0	9.6-9.9	2.6	5.2-5.8	90	0.4
Encapsulation in 74S/26AN	20	16-17	1.7	11-12	105	2.5
Encapsulation in 72S/26AN/ 2MAA	20	16.5-18	1.8-1.9	11-12	107	1.7-2.3
Dry blends with SAN <sup>a</sup>	20	14-14.5	1.6	10.8-11.3	103-105	2.2-2.5
Commercial blend <sup>b</sup>	20	13-14	1.3	10.3-11.3	103	2.1

<sup>a</sup> Copolymer of 74S/26AN.

<sup>b</sup> Commercial blend of SAN and long glass concentrate.

TABLE IV  
 Properties of Composites of Glass and SAN Prepared by the Encapsulation Process  
 (All Composites Contain 20% by Weight of Glass Strands—Extruded and Injection Molded)

Composition of polymer used in encapsulation	% Glass in original capsules	Glass size, in.	Diluent	Tensile, psi $\times 10^{-3}$	% elongation at fail	Tensile modulus, psi $\times 10^{-5}$	Izod impact $\frac{1}{2} \times \frac{1}{8}$ in., ft-lb/in.	HDT <sup>a</sup> at 264 psi, °C
74S/26AN	20	$\frac{1}{8}$	none	15-16.5	1.7	10.5-11.5	2.3	103-106
74S/26AN	71	$\frac{1}{8}$	SAN <sup>b</sup>	17.2	1.8	12.0	2.3	108
74S/26AN	54	$\frac{1}{4}$	SAN	16.1	1.8	11.3	2.7	105
72S/26AN/2MAA	65	$\frac{1}{8}$	SAN	17.6	1.9	11.6	2.3	107
72S/26AN/2MAA	77	$\frac{1}{8}$	SAN	17.4	2.0	11.6	1.7	105
72S/26AN/2MAA	65	$\frac{1}{4}$	SAN	16.6	1.8	11.0	2.7	107

<sup>a</sup> Heat distortion temperature.

<sup>b</sup> 74S/26AN.

TABLE V  
Composites Prepared by Encapsulation of 20% (by Weight) of  $\frac{1}{8}$ -in. Glass Strands in 74S/26AN

Protective colloid	Glass/mon. charge	Tensile strength, psi $\times 10^{-3}$		% Elongation		Modulus $\times 10^{-5}$		Izod impact, <sup>c</sup> ft-lb/in.		
		IM <sup>a</sup>	EXT + IM <sup>b</sup>	A <sup>c</sup>	IM	EXT + IM	A		IM	A
B	$\frac{1}{4}$	15-15.5	15-15.7	15.9	1.7	1.7	11.5-12.5	11.5-12.5	11.3	2.3
A	$\frac{1}{4}$	15.5-16	15.5	16.6	1.6	1.8	11.5-11.8	10.5-11.4	11.4	3.2
Commercial blend (20% glass)			13-14			1.3		10.3-11.6		2.1

<sup>a</sup> Injection molded directly without extrusion in 1-oz machine.

<sup>b</sup> Extruded at 450°F with 1-in. vinyl screw. The extruded strands are then injection molded (IM) in 1-oz machine.

<sup>c</sup> Molded in Arburg machine with SAN screw.

TABLE VI  
Properties of Composites Containing 20 Weight % Glass Strands (Extruded and Injection Molded)

Composition of polymer used in encapsulation <sup>a</sup>	SAN, <sup>b</sup> % diluent,	Tensile, psi $\times 10^{-3}$		Elongation at fail		Tensile modulus, psi $\times 10^{-6}$		Izod impact, ft-lb/in. $\frac{1}{2} \times \frac{1}{8}$ in., Dry	Heat distortion at 264 psi, °C Dry	
		Dry <sup>c</sup>	Wet <sup>d</sup>	% Loss	Dry	Wet	Dry			Wet
74S/26AN	none	16.6	15.4	8	1.8	1.7	11.4	10.9	3.2	103-106
74S/26AN	90	17.2	16.2	6	1.8	1.7	12.1	12.0	2.3	108
72S/26AN/2MAA	82	17.3	16.7	4	1.8	1.7	11.5	11.5	2.2	107
72S/26AN/2MAA	86	17.2	16.4	5	1.7	1.7	12.6	12.1	---	106
71S/25AN/4MAA	83	16.4	16.5	0	1.6	1.7	12.1	11.5	2.0	107
Dry blend (no encapsulation)	100	14.7	13.0	13	1.8	1.5	10.8	10.2	2.1	104
Commercial blend		13.8	12.0	15	1.3	1.3	11.6	10.4	2.1	103

<sup>a</sup> MAA = Methacrylic acid.

<sup>b</sup> Amount of SAN, expressed in % on total matrix (i.e., % on SAN diluent + copolymer in capsules), used to dilute the capsules to 20% glass.

<sup>c</sup> Dry = Standard 50% humidity.

<sup>d</sup> Wet = Specimens exposed to 80% humidity at 60°C for 42 hr, then heated at 80-85°C in water for 24 hr, then air dried.

probably be attributed to the preferential orientation of fibers in the machine direction observed in injection molded specimens. Burning away of the matrix from injection-molded specimens, while leaving the fibers undisturbed, does in fact reveal preferential orientation in the machine direction, but a large number of fibers are found oriented at random and perpendicular to the machine direction.

Composites prepared from encapsulated glass are found, by qualitative microscopic examination, to preserve much greater glass integrity than composites obtained from dry-blends of glass strands and a preformed SAN matrix. In addition, a considerably lower loss in properties is experienced (Table VI) on prolonged exposure to water.

Water resistance of glass composites is generally considered poor because of the glass fibers behaving as wicks and adsorbing water by capillary action with resulting damage to the glass-matrix adhesive bond and to mechanical properties. Microcracks are thus formed along the glass fibers which scatter light and reduce the translucency of the molding. This undesirable behavior is much less evident with composites obtained by encapsulation.

The improved mechanical properties and water resistance obtained with composites prepared by encapsulation are probably due to one or more of the following factors:

a. Good wetting by adsorption of monomers by the strands and by the fibrils within the strands. This is possible because of the low viscosity of the monomers compared to the more difficult wetting and penetration by a viscous polymer melt.

b. The monomers are believed to improve matrix-to-strand adhesion by dissolving or swelling the glass size during polymerization. Grafting of the size, or reaction with the silanes, may also take place.

c. The presence of polymer on the fibrils and the glass strands results in reduced abrasion of glass against glass, and thus reduced glass damage, on processing.

There is no apparent gain in properties (Table IV), except for a small gain in impact strength, by using  $1/4$ -in. in place of  $1/8$ -in. strands in encapsulation.

Glass concentrates containing more than 50% by weight of glass fibers can readily be prepared by the encapsulation process (Table II). Their dilution to 20% glass with preformed SAN results in composites of better properties (Tables IV and VI) than those obtained by direct encapsulation of 20% glass in SAN. No obvious explanation is available for this behavior.

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