DEC 30 1964

MASTER

ADVANTAGES OF GLASS MICROBALLOONS

AS FILLER MATERIAL

August 10, 1960

Prepared by

F. R. Walterbach

Publication Unknown

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission, makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe upon any rights or trademarks.

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of, any information, apparatus, method, or process disclosed in this report.

A. Used in the above, "person" acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

PATENT CLEARANCE OBTAINED. RELEASE TO
THE PUBLIC IS APPROVED. PROCEDURES
ARE ON FILE IN THE RECEIVING SECTION.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

ADVANTAGES OF GLASS MICROBALLOONS AS FILLER MATERIAL

By F. R. Walterbach, Materials Engineering Dept., The Bendix Corporation, Kansas City Division

As the environmental requirements for electronic equipment have become increasingly severe, it has become necessary for designers to protect their circuits by encapsulating them in Plastic Resins. In a well-designed system this plastic encapsulation almost completely isolates the equipment from the detrimental effects of humidity and mechanical shock.

The many configurations and varied requirements of electronic equipment complicate the choice of resin system for any particular application, particularly as the manufacturing characteristics must also be considered. At the present time epoxies are the most common plastic resin in use. The epoxy resin usually contains a large quantity of some filler material such as finely divided silica, calcium carbonate, or mica. Designers generally prefer to use the maximum amount of filler which can be easily processed, since resistance to thermal shock is usually increased with more filler. One of the problems associated with potting compounds is the difference between the thermal coefficient of expansion of the potting compound and that of the components being potted. Large quantities of inorganic filler lower the thermal coefficient to more nearly match the expansion of associated metals and ceramics. Fillers also serve to reduce cure shrinkage, increase thermal conductivity, lower the curing exotherm, and to extend the pot life during fabrication.

One of the drawbacks of an encapsulated assembly is the increase in weight of the assembly, caused by the potting compound replacing the air surrounding the individual components. Since many of these

encapsulated assemblies are airborne, it is desirable for the weight of the assembly to be reduced to the minimum. Most of the encapsulating systems have specific gravities in the range of 1.2 to 1.8. The reduction of these figures involved only the finding of a suitable filler which has a lower specific gravity. The lowest specific gravity yet found is with microballoons made from phenolic, urea-formaldehyde, or glass compounds. These microballoons are minute (30-300 micron diameter) hollow spheres.

When The Bendix Corporation (Kansas City Division) began to use these filler materials the first impression was favorable, but inconsistencies developed later in the thermal shock resistance tests. To better understand this characteristic an investigative program was initiated, to determine the ideal particle size and specific gravity to produce a filled epoxy with optimum thermal shock properties. Filler materials were combined with epoxy resin and the following tests were made to determine specific gravity, thermal conduction, coefficient of expansion, thermal shock, tensile modulus at room and low temperature, dielectric constant and dissipation factor.

Particle Size Distribution

To separate the microballoons according to particle size, a Cenco Meinzer shaker with standard 60, 100 and 200 mesh screens was used. The sieve shaker with a load of 100 grams was allowed to vibrate until the material on the bottom screen remained at constant weight. After several checks, twenty minutes proved to be the time necessary to accurately separate the material. This method gave five particle size distributions:

1. Material collected on the 60 mesh screen.
2. Material collected on the 100 mesh screen.
3. Material collected on the 200 mesh screen.
4. Material passing through the 200 mesh screen.
5. Material as received in the shipping containers (that was not fractioned according to particle size or specific gravity).

The material collected on the 60 mesh screen was of a negligible amount and will be disregarded at this point (see Table 1 for an average sieve analysis).

TABLE 1
PARTICLE SIZE DISTRIBUTION

<u>Particle Size</u>	<u>Percent of Total</u>
Material collected on 60 mesh screen	0-1
Material collected on 100 mesh screen	5-20
Material collected on 200 mesh screen	40-60
Material passed through 200 mesh screen	25-60

The different fractions from above were then placed in acetone (specific gravity 0.792) and separated according to whether they floated or sank (see Table 2). Any material not floating on top of the acetone was considered to be part of the fraction that had sunk.

TABLE 2
ACETONE FLOTATION RATIOS

<u>Particle Size</u>	<u>Percent Submerged</u>	<u>Percent Floating</u>
Collected on 100 mesh screen	20-50	50-80
Collected on 200 mesh screen	15-36	64-85
Passed through 200 mesh screen	35-40	60-65
As received in shipping containers	20-35	65-80

This now made available the eight samples of filler outlined above and material from each screen that had not been placed in acetone, giving a total of twelve samples (microphotographs of each of these samples are pictured in Figure 1).

{Insert Figure 1 here}

Specimen Preparation

The filler material was combined with an epoxy resin (Epon 828) manufactured by the Shell Chemical Co., in 25-75 and 30-70 ratios. It was found that when the filler resin ratio was 30-70 the larger microballoons lighter than acetone rendered the mix too viscous for conventional potting compounds. Therefore, all microballoons lighter than acetone were combined in a 22-78 ratio, and test specimens were prepared as outlined below:

1. Balloons and resin were mixed in proper proportions in approximately 1000-gram batches for 15 minutes, at No. 2 speed on a "Kitchen - Aid" Hobart-type mixer.
2. Preheated for 30 minutes at $160 \pm 5^{\circ}\text{F}$
3. Diethanolamine (curing agent) added (12 parts per 100 parts of resin)
4. Mixed for 3 minutes by hand
5. Returned to oven for 10 minutes from time of hardener addition
6. Vacuum pulled on mix three minutes after foam broke at 1-3 mm
7. Samples poured
8. Cured at $155 \pm 5^{\circ}\text{F}$ for 24 hours.

All test specimens were poured in oversize open molds and machined to size. A portable DK analyzer was used to take readings on thermal conduction specimens for dielectric constant and dissipation factor measurements.

The following conclusions were drawn from the microphotographs of Figure 1:

1. Sieving breaks a few microballoons.
2. Sieving provides a sharp separation into the expected particle sizes.

3. Low density fraction (that floated in acetone):
 - a. Tends to have approximately 5 per cent sinkers
 - b. Is mostly perfectly spherical (floaters)
 - c. Not sieved is heavily agglomerated.
4. High-density fraction (that sank in acetone):
 - a. Consists of solid spheres, porous glass or broken oversize balloons
 - b. Of smaller particle size is composed of thick-walled and solid spheres.

All tabulated data shown in Table 3 is an average figure of test specimens. The results were consistent except for dielectric constant readings (and the figures on the different tests for the 22-78 filler epoxy mix). The specimens for the dielectric readings were 1/8 - inch minimum thickness, with a 10 percent average deviation in readings. The correlation between dielectric constant and specific gravity is shown in Figure 2.

{Insert Figure 2 here}

(TABLE 3)

The specific gravity of the cured mix was in direct proportion to the particle size specific gravity of the filler and the ratio of resin to filler. For example, the lightest mix possible, although too viscous to pour, would have been particles on the 60 mesh screen that floated in acetone and were combined in a 30-70 ratio. This mix would have been the lightest material possible in this experiment.

Thermal shock specimens were prepared by suspending a standard 1 - inch pipe 2 inches long in an 8 ounce cup and pouring the mix around it. A cross - sectioned sample is pictured in Figure 3. The sample was then exposed to -65°F for 4 hours and brought back to room temperature; this cycle was repeated 10 times.

(Insert Figure 3 here)

The sample pictured cracked in every way possible. The crack around the inside of the pipe appeared to be caused by mix pulling away from the pipe. This break only extended down to the edge of the pipe and did not go beyond. In a mix that had low density filler (lighter than acetone) and failed, this is the predominant type of crack that appeared.

Both types of cracks were in evidence for the high density material. The outside crack is believed to be caused by a combination of too high a tensile modulus and coefficient of expansion. Sometimes one thermal shock sample cracked while the other two did not. Several explanations may be given for this. Either there was a sharp corner or burr that hadn't been machined off the embedded pipe or the other two samples were near the cracking point.

The tensile modulus was checked at -65°F and room temperature to give an indication as to how thermal shock would be affected; this property was dependent on the specific gravity of the filler and the size of the filler. A low specific gravity and a large particle size gave a low tensile modulus; this was expected due to the greater volume of filler in the composition.

Thermal conduction specimens were the only ones that did not give the expected results, as greater percentages of glass increased the conduction. It was thought that unbroken spheres of low density containing "dead" air would act as insulators, but it appears that air space is a negligible factor.

Conclusive Advantages

The tests showed that microballoons have approximately the same thermal conduction as a mix without filler. This makes the balloons more attractive as a filler for two reasons: a low coefficient of

expansion is available, and thermal conduction is decreased a negligible amount compared to an unfilled epoxy.

No correlation was found between coefficient of expansion and specific gravity in epoxy resin-glass microballoon mixes until a particular microballoon particle size was held constant. Figures 4 and 5 are illustrations of this, showing the curves independent of the ratio of resin to filler. The same requirement of constant particle size was valid for factors of tensile modulus and thermal conduction.

A variation in particle size is desirable, as the microballoon filler disperses more evenly throughout the resin. An area with only resin will crack under thermal shock much more easily than one containing filler.

(Insert Figures 4 and 5 here)

BDX-613-6

TABLE 3
TEST SPECIMEN TABULATION

	Specific Gravity	Coeff. of expansion x 10 ⁶ in/in/°F -65° to 78° F 78° to 165° F		Thermal Conduction 10 ⁴ x $\frac{\text{Cal-Cm}}{\text{Sec-Cm}^2-\text{°C}}$	Tensile modulus x10 ⁻⁵ psi -65° 78°		Thermal shock 10 cycles	Dielectric Constant	Dissipation Factor D x 10 ⁴ @10kc
+ 100 mesh									
70 - 30 ratio Heavy Light	Not enough material The mix was too viscous for any samples to be poured								
75 - 25 ratio Heavy	1.28	20.8	28.3	5.04	7.1	--	3 failed 1st cycle	4.00	2.40
Light	0.702	--	--	4.56	--	--	3 passed	2.90	2.1
+ 200 mesh									
70 - 30 ratio Heavy Light	1.37 Too viscous	18.8	26.8	5.22	9.1	6.3	3 failed 1st cycle	4.15	2.15
75 - 25 ratio Heavy	1.23	25.4	31.7	4.94	7.4	5.0	2 failed 1st cycle 1 passed	4.20	2.25
Light	0.765	17.9	20.9	4.47	4.6	3.9	3 passed	2.60	2.10
- 200 mesh									
70 - 30 ratio Heavy Light	1.23 0.851	22.1 16.7	28.5 22.3	5.55 4.89	7.5 5.2	5.7 4.7	3 passed 3 passed	4.35 3.45	2.45 2.20
75 - 25 ratio Heavy	1.22	21.1	33.1	4.90	7.3	6.0	2 failed 1st cycle 1 failed 3rd cycle	4.50	2.20
Light	0.887	18.3	24.2	4.76	5.7	4.6	3 passed	3.40	2.55
As Received									
70 - 30 ratio Heavy	1.337	19.2	27.2	5.23	--	--	3 failed 1st cycle	4.60	1.90
Light	0.753	22.5	23.1	4.39	10.0	--	3 passed	2.70	2.00
75 - 25 ratio Heavy	1.20	21.5	27.2	4.88	12.1	6.9	3 failed 1st cycle	4.20	2.00

TABLE 3 (Cont.)
TEST SPECIMEN TABULATION

	Specific Gravity	Coeff. of expansion x 10 ⁶ in/in/°F -65° to 78° F 78° to 165° F		Thermal Conduction 10 ⁴ x $\frac{\text{Cal-Cm}}{\text{Sec-Cm}^2 \cdot ^\circ\text{C}}$	Tensile modulus x10 ⁻⁵ psi -65° 78°		Thermal shock 10 cycles	Dielectric Constant	Dissipation Factor D x 10 ⁴ @10kc
75 - 25 ratio Light	0.810	16.8	20.0	4.65	5.9	4.25	1 failed 2nd cycle 2 passed	3.00	2.05
70 - 20 ratio									
As Received, light									
+ 100, Light	0.867	17.7	25.2	4.95	4.8	4.4	3 passed	2.9	1.90
+ 200, Light	0.755	16.8	23.1	4.60	4.7	4.1	3 passed	2.8	2.20
+ 200, Light	0.785	18.2	21.5	5.03	5.0	4.7	3 passed	3.0	1.70
- 200, Light	0.919	17.8	26.0	5.33	5.6	4.7	3 passed	3.2	2.00
+ 100 mesh - No separation in Acetone									
70 - 30 ratio	0.850	--	--	5.13	--	4.7	3 passed	3.05	2.50
75 - 25 ratio	0.931	17.7	22.7	5.01	7.9	4.0	3 passed	3.55	2.05
+ 200 mesh - No Separation in Acetone									
70 - 30 ratio	0.923	18.9	24.5	5.10	5.6	4.7	1 failed 2nd cycle 2 passed	3.05	2.50
75 - 25 ratio	0.941	18.0	22.4	4.93	5.3	4.1	3 passed	3.30	4.00
- 200 mesh - No Separation in Acetone									
70 - 30 ratio	1.03	20.3	24.4	7.99	9.7	4.8	3 passed	3.30	2.10
75 - 25 ratio	0.970	19.0	25.2	5.24	7.3	4.8	3 passed	3.50	2.25
As Received - No Separation in Acetone									
70 - 30 ratio	0.846	16.4	22.2	5.39	5.4	4.9	3 passed	3.35	2.65
75 - 25 ratio	0.942	17.3	20.4	5.06	5.5	4.2	3 passed	3.20	2.50

a.

b.

- a. Material as it is received in shipping container.
- b. Material from "a" that sank in the acetone separation.
- c. Material from "a" that floated in the acetone separation.

c.

Figure 1. Microphotographs of Glass Microballoons, 60X Magnification.

d.

e.

f.

- d. Material collected on 100 mesh screen.
- e. Material collected on 100 mesh screen, that sank
- f. Material collected on 100 mesh screen, that floated in acetone.

Figure 1. Microphotographs of Glass Microballoons, 60X Magnification. (continued)

g.

h.

g. Material collected on 200 mesh screen.

h. Material collected on 200 mesh screen, that sank acetone.

j. Material collected on 200 mesh screen, that floated in acetone.

j.

Figure 1. Microphotographs of Glass Microballoons, 60X Magnification, (continued)

k.

l.

m.

- k. Material passed through a 200 mesh screen.
- l. Material passed through a 200 mesh screen, then sank in acetone.
- m. Material passed through a 200 mesh screen, then floated in acetone.

Figure 1. Microphotographs of Glass Microballoons, 60X Magnification. (continued)

Figure 2. Thermal Shock Specimen.

DIELECTRIC CONSTANT
VS
SPECIFIC GRAVITY

Figure 3.

PARTICLE SIZE HELD CONSTANT
THROUGH 100 MESH,
CAUGHT ON 200 MESH

Figure 4.

PARTICLE SIZE HELD CONSTANT

- 200 Mesh

Figure 5.