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METHOD FOR MAKING A SOLID DIELECTRIC
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tion of Delaware
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This invention relates to dielectric materials employed in the manufacture of articles adapted to control the passage of electrical energy and more particularly to the composition of a dielectric material having a uniform dielectric constant and the method of making the same. The dielectric constant of a material is defined as the ratio of the velocity of propagation of the electrical energy through the material relative to a vacuum.

Uniformity of the dielectric constant of materials of the type referred to above, which determines the operating efficiency of the end article produced therefrom, has heretofore never been satisfactorily obtained in so-called "foamed" dielectric materials. In "foamed" dielectric materials the globules forming the material are mass-made simultaneously while the material is being produced. Therefore, where uniformity has been obtained, it has been through the use of so-called "syntactic foams." As opposed to "foamed" dielectrics, "syntactic foams" are the result of comparatively complex and expensive manufacturing processes involving the preforming of individual globules in a variety of sizes for different applications and classifying the several sizes to the end that a number of globules of a selected size may be subsequently bonded together to form the ultimate material or article. In order to avoid these complicated manufacturing processes, "foamed" dielectric materials have been resorted to, particularly in large scale production.

At the same time, however, articles such as, for example, lenses, radomes, etc., made of existing foamed materials effect relatively erratic deflections and refractions of impinging electrical energy which they are intended to control and/or focus. The present invention is, therefore, directed to an improved foamed dielectric material having a uniform dielectric constant to the end that the direction of the electrical energy may be accurately and readily controlled. In addition, following the teachings of the instant invention, the dielectric constant of the material may be varied, i.e., adjusted, whereby the ultimate article may be made to more closely correspond to, for coaction with, a particular associated piece of equipment in a given application. Hence, the operating efficiency of the end article is improved over articles made from prior foamed dielectric materials.

More specifically, the invention contemplates a foamed dielectric material comprising specific ingredients so combined as to produce unitary globules that are substantially identical one to the other in size and composition so that the ultimate article produced therefrom is more homogeneous in structure and its function improved. These globules may also, when desired, be made relatively small whereby substantial strength, relative to frangibility and thermal resistance, is obtained in the end article. Strength of the dielectric material is important in certain applications for which the end article is designed, e.g., a dielectric lens in present day aircraft is often subjected to temperatures which can and do cause a failure or operational breakdown of the lens.

With the above and other objects in view, as will be apparent, this invention consists in the preparation of two separate and distinct mixtures, each under specific conditions and subsequently combined to produce an ultimate mix from which the foamed dielectric material contemplated herein is made. Broadly, this practice has been followed heretofore in producing foamed dielectric

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materials. However, the ingredients, as well as the conditions under which the ultimate mix is obtained, differ in several critical respects from known practices.

As opposed to prior practices, the first mixture, Mix No. 1 herein, employs a base or binder material of epoxy resin falling within a relatively high melting point range, i.e., between 40° and 75° C. (104° and 167° F.), and preferably having a viscosity at 25° C. of between .3 and 1.6 poises taken in a 40% weight solution of butyl Carbitol. One such epoxy resin is presently produced and sold by the Shell Chemical Company under the trade name of "Epon 836."

With this base or binder is mixed, at a temperature above the melting point of the particular epoxy used, a filler of materials having high mechanical and conductive properties. The quantities of these filler materials used determines the ultimate strength and dielectric constant, respectively, of the ultimate material. In specific cases where the strength of the ultimate article is not important and a relatively low dielectric constant (in the order of 1) is desired, both of these fillers may be omitted. On the other hand, where either a relatively strong or a relatively high dielectric constant alone is desired, then only the corresponding filler is used. However, in the majority of cases, some of each of these fillers is required. When employed, up to 50 parts by weight of the filler with the high mechanical properties, e.g., milled glass, slate, etc., may be used for every 100 parts by weight of epoxy resin. Up to 30 parts by weight of the filler with the high conductive properties, preferably metal such as aluminum powder, may be used for every 100 parts by weight of epoxy resin.

When either or both of the foregoing fillers are employed, a suspension agent or soap, preferably a metallic salt of an organic acid such as aluminum stearate, is necessary in the mixture. Depending upon the particular amount of filler used, up to 6 parts by weight of the suspension agent may be used for every 100 parts by weight of epoxy resin. It is the function of the suspension agent to prevent the particles comprising the fillers from settling in the liquid epoxy resin during the subsequent foaming and curing operation to the end that they are scattered throughout and separated one from the other by the resin to produce a homogeneous material.

Whether or not a filler and suspension agent is used, the invention further contemplates a flame-retardant additive to Mix No. 1, to the end that the ultimate article will not be prone to support combustion. Several such additives, e.g., antimony oxide, chlorinated wax, etc., or a compound of these, may be employed for this purpose, it being the function of the additive upon the application of heat to produce a flame-smothering gas. Between 2 and 20 parts by weight of the additive or a combination of additives is used for every 100 parts by weight of epoxy resin.

The second mixture, Mix No. 2, which is separately and independently prepared, as above stated, comprises a foaming or blowing agent that includes an amine, e.g., dinitrosopentamethylenetetramine, diazo amino benzene, ammonium carbonate, etc., mixed at room temperature with a curing agent that includes a liquid aromatic amine or eutectic mixture of aromatic amines, e.g., metaphenylene diamine 4, 4' methylene dianiline; 2,6-diamino pyridine; 4 chloro-ortho-phenylene diamine; diamino diphenyl sulfone, etc. E. I. duPont de Nemours and Company manufactures and sells such a blowing agent under the trade name of "Unicel N.D." An example of a ready-made curing agent of the type specified above that is readily available on the market is "Curing Agent Z" produced by the Shell Chemical Company.

For every 100 parts by weight of epoxy resin used in Mix No. 1 with which Mix No. 2 is to be subsequently

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blended, from a fractional part up to 6 parts by weight each of the curing agent and blowing agent is required.

While not essential, in many cases depending upon the particular combination and quantities of ingredients used in Mix No. 1 and Mix No. 2, an additive is blended into Mix No. 2 to disperse or accelerate the dispersion of the foaming agent. The quantity of such an additive included depends upon and is in proportion to the quantity of foaming agent used. Thus, up to 6 parts by weight of the additive may be employed for every 100 parts of epoxy resin. This additive or dispersion agent is any epoxy resin that is liquid at room temperature, preferably one having a viscosity at 25° C. of from 40 to 100 poises. One such dispersion agent available on the market is "Epon 820" produced and sold by the Shell Chemical Company.

When Mixes No. 1 and 2 have thus been produced, they are combined one with the other in the following manner. While Mix No. 1 is maintained at an elevated temperature, i.e., a temperature above the melting point of the particular epoxy resin, Mix. No. 2 is added thereto and mixed therewith, as by stirring, until the temperature of the resultant mix reaches approximately 200° F. This rise in temperature is the result of the curing action which occurs.

The resultant mix while at a temperature of about 200° F. may thereafter be poured into a mold corresponding to and defining the shape of the ultimate article and which has been saturated with a temperature of about 250° F. With the resultant mix in the heated mold, a temperature of approximately 250° F. is applied to and maintained against the mold until the resultant mix is fully foamed and cured. This requires from about 9 to 14 hours, depending upon the amount and composition of the particular mixture. The foamed dielectric material having thus been produced, it is thereafter removed from the mold and worked or shaped, if desired, by conventional mechanical means well known to the art, to the finished article.

Referring by way of example to a specific batch of foamed dielectric material to be made into a lens for the guidance system of a relatively high altitude, high speed airplane designed to operate at approximately 9800 megacycles, a base of epoxy resin with a melting point at 40° to 45° C. is heated to approximately 235° F. For every 100 parts by weight of epoxy resin, approximately 20 parts of milled glass fiber, 15 parts of aluminum powder, 2 parts aluminum stearate and 10 parts each of antimony oxide and chlorinated wax, all at room temperature, are added to the heated epoxy resin. The temperature of this mixture is raised to and maintained at about 235° F. after which the mixture is stirred or otherwise blended for approximately two hours. The mixture is then removed from the heat and blending is continued while the temperature thereof drops to about 180° F. This constitutes Mix No. 1.

Separately, Mix No. 2 is prepared by mixing for every 100 parts by weight of the epoxy resin 4 parts by weight each of the curing agent, the foaming agent and the dispersion agent at room temperature. This mixture, Mix No. 2, is added to Mix No. 1 while Mix No. 1 is at approximately 180° F. and being stirred or blended. This stirring is continued until the combination of mixes reaches a temperature of about 200° F. due to the curing action whereby the ultimate mix is obtained.

During the foregoing operations, the mold of the lens is placed in an oven or otherwise heated to a temperature of about 250° F. for at least two hours. When the mold has thus been heat-soaked, and while the ultimate mix aforesaid is at a temperature of about 200° F., the ultimate mix is poured into the mold. The mold is maintained at a temperature of approximately 250° F., preferably in an oven, until the dielectric material is fully foamed and cured. As stated, this takes from approxi-

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mately 9 to 14 hours depending on the quantity and specific composition of material employed.

The ultimate lens produced in this particular case would be comprised of a homogeneous dielectric material having a dielectric constant of about 3.0 at 9800 megacycles, a density of about 14.85 lbs. per cubic foot and an average compressive strength of about 800 p.s.i. These results may be repeatedly obtained an unlimited number of times without substantial variation whereby lenses having substantially identical characteristics may be produced.

What is claimed is:

1. The method of making a cured resinous foam adapted to be molded into a lens having a predetermined dielectric constant corresponding to associated electrical energy-transmitting and receiving equipment consisting essentially of mixing with every 100 parts by weight of an epoxy resin falling within a melting point range of 40° to 75° C. up to 50 parts by weight of ceramic particles, up to 30 parts by weight of metallic particles and up to 6 parts by weight of a polyvalent metal soap suspension agent at an elevated temperature above the melting point of the epoxy resin whereby said particles are scattered throughout and separated one from the other by the resin, separately mixing at room temperature for every 100 parts by weight of the epoxy resin from a fractional part up to 6 parts by weight of a liquid aromatic amine curing agent with from a fractional part up to 6 parts by weight of an amine soap whereby a foaming reaction occurs, combining the separate mixes aforesaid at said elevated temperature prior to the commencement of said foaming reaction, and applying additional heat to the combined mixes until fully foamed and cured.

2. The method of making a cured resinous foam material having a selected range of dielectric constants to effect the refraction of electrical energy passing therethrough at predetermined angles corresponding to associated electrical energy-transmitting and -receiving equipment consisting essentially of mixing with every 100 parts by weight of a base epoxy resin falling within a melting point range of 40° to 75° from a fractional part up to 30 parts by weight of metallic particles and from a fractional part up to 6 parts by weight of a polyvalent metal soap suspension agent at a temperature within the melting point range aforesaid, separately mixing at room temperature for every 100 parts by weight of the epoxy resin from a fractional part up to 6 parts by weight of a liquid aromatic amine curing agent with a fractional part up to 6 parts by weight of an amine forming agent, combining the separate mixes aforesaid at a temperature within the melting point range aforesaid, applying additional heat up to approximately 250° F. to the combined mixes, and maintaining said combined mixes at such additional temperature until fully foamed and cured.

3. The method of making a cured resinous foam having a range of dielectric constants to effect the refraction of electrical energy passing therethrough at effective predetermined angles corresponding to associated electrical energy-transmitting and -receiving equipment consisting essentially of mixing with every 100 parts by weight of a base epoxy resin falling within a melting point range of 40° to 75° C. and having a viscosity at 25° C. of between .3 and 1.6 poises taken in a 40% weight solution of diethylene glycol monobutyl ether from a fractional part up to 30 parts by weight of metallic particles and from a fractional part up to 6 parts by weight of a polyvalent metal soap suspension agent at a temperature within the melting point range aforesaid, separately mixing at room temperature for every 100 parts by weight of the epoxy resin from a fractional part up to 6 parts by weight of a liquid aromatic amine curing agent with from a fractional part up to 6 parts by weight of an amine foaming agent, combining the separate mixes aforesaid at a temperature within the melting point range aforesaid, applying additional heat up to approximately 250° F. to the

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combined mixes, and maintaining said combined mixes at such additional temperature for approximately from 9 to 14 hours.

4. The method of making a cured resinous foam having a preselected dielectric constant consisting essentially of mixing with every 100 parts by weight of an epoxy resin falling within a melting point range of 40° to 75° C. up to 50 parts by weight of particles of milled glass, from a fractional part up to 30 parts by weight of powdered aluminum and from a fractional part up to 6 parts by weight of a polyvalent metal soap at a temperature above the melting point of the resin whereby said particles are scattered throughout and separated one from the other by the resin, separately mixing at room temperature for every 100 parts by weight of the epoxy resin from a fractional part up to 6 parts by weight of a liquid aromatic amine curing agent and from a fractional part up to 6 parts by weight of an amine foaming agent, combining the separate mixes at the temperature above the melting point temperature aforesaid, and applying additional heat to the combined mixes until fully foamed and cured.

5. The method of making a cured resinous foam having a preselected dielectric constant consisting essentially of preparing a first mixture at a temperature within a range of 40° and 75° C., of an epoxy resin having a melting point within said temperature range with for every 100 parts of the resin used up to 50 parts by weight of ceramic particles, from a fractional part up to 30 parts by weight of metallic particles, from a fractional part up to 6 parts by weight of a suspension agent of a polyvalent metal soap and between 2 and 20 parts by weight of a flame-retardant material, separately preparing a second mixture at room temperature of from a fractional part up to 6 parts by weight each of an amine foaming agent and a liquid aromatic amine curing agent for every 100 parts by weight of the resin, combining said first and second mixtures into a resultant mix while the first mixture and the resultant mix is maintained at the mixing temperature of the first mixture aforesaid, elevating the resultant mix to a temperature in the order of 250° F., and main-

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taining said resultant mix at such elevated temperature until fully foamed and cured.

6. The method of making a cured resinous foam material adapted to be molded into a lens having a dielectric constant of about 3.0 at 9800 megacycles, a density of about 14.85 lbs. per cubic foot and an average compressive strength of about 800 p.s.i. consisting essentially of mixing with every 100 parts by weight of a base epoxy resin having a melting point at about 40° to 45° C. heated to about 235° F. approximately 20 parts by weight of milled glass fiber, 15 parts by weight of aluminum powder, 2 parts by weight aluminum stearate and 10 parts by weight each of antimony oxide and chlorinated wax all at room temperature, heating this mixture to and maintaining it at about 235° F. and blending it for approximately 2 hours, continuing to blend the mixture while reducing its temperature to about 180° F., separately mixing at room temperature for every 100 parts by weight of the epoxy resin about 4 parts by weight each of a liquid aromatic amine curing agent, an amine foaming agent and a dispersion agent of epoxy resin liquid at room temperature, combining and blending the separate mixes aforesaid at a temperature of approximately 180° F., and elevating the heat of the combined mixes to approximately 200° F. while they are being blended.

References Cited in the file of this patent

UNITED STATES PATENTS

30	2,739,134	Parry et al	Mar. 20, 1956
	2,797,201	Veatch et al	June 25, 1957
	2,806,509	Bozzacco et al	Sept. 17, 1957
	2,822,448	Wegmann	Feb. 4, 1958
	2,831,820	Aase et al	Apr. 22, 1958
35	2,912,382	Liao et al	Nov. 10, 1959

OTHER REFERENCES

- The Merck Index, Merck & Co., Rahway, N.J., 1952, 6th ed. page 86.
- 40 Lee et al.: Epoxy Resins, McGraw Hill, New York, 1957, pages 148-151, 188, 203-211, 281.