

opaque fused quartz may be used. The latter is cheaper and may lead also to a desirable slight decrease in density when substituted for the clear material in a given preparatory process.

The use of fused quartz as the sole component in the production of silica spheroids would lead to the development of a product with the density of vitreous silica, about 2.2 grams per cubic centimeter. A second component, silica gel, is, in this invention, mixed with the fused quartz to bring about a modification of the density.

Silica gel is a form of silicon dioxide which is extremely porous and which contains water chemically bound and trapped within the porous structure. The density of silica gel is about 2.0 grams per cubic centimeter. If the gel is expanded according to the method taught in the Christensen et al. Patent, No. 2,151,083, mentioned earlier, a material with a density about one-half to one-third of this value is obtained.

Silica gel, which may be obtained by acid-induced precipitation of silica from solutions of soluble silicates, is thought to have a variable amount of water associated with the silicon dioxide therein. Release of this water, during the spheroidizing taught in the patent referred to, causes tumescence or puffing of the silica, and leads to fused material of the very low densities mentioned.

Not all the water in silica gel is bound with equal tenacity in the porous structure of this material however. It has been found possible, by a firing at temperatures below those found in an atomic hydrogen arc, to remove substantial portions of the more-loosely bound water. The remaining water in the structure will then form voids upon spheroidizing which are more homogeneously distributed throughout the fused material. The voids, further, are of smaller size, and have less tendency to break through the sphere wall. Finally, better control over void formation can be obtained by this process of driving off indeterminate and variable amounts of loosely-bound water in a firing preliminary to spheroidizing. A more constant amount of tightly-bound water remains in the gel. This more uniform water content leads to greater reproducibility and uniformity of the product.

More importantly, it has been discovered that the density of fused silica spheroids produced by expansion in an atomic hydrogen arc can be determined by using a mixture of fused quartz, as a high-density component, and silica gel, as the material furnishing the water vapor responsible for the expansion. The higher the ratio of fused quartz to silica gel, the closer the approach of the product's density value to 2.2 grams per cubic centimeter, the density of fused quartz or vitreous silica. The lower the ratio of fused quartz to silica gel, the more closely do the product densities approach those of the buoyant structures taught by Christensen et al. in the Patent No. 2,151,038. By varying the ratio at intermediate values, fused silica spheroids with densities between the values of approximately 0.05 gram per cubic centimeter and 2.2 grams per cubic centimeter can be readily obtained.

The third component of the forming mixture described herein, colloidal silica, is used as a binder in the fused quartz-silica gel mixture. Since the colloidal silica functions primarily as a non-contaminating binder for the other ingredients, its proportion in the forming mixtures remains essentially constant, though the ratio of fused quartz to silica gel is varied.

The compositions found most advantageous for obtaining the density of 1.5 grams per cubic centimeter desired in microphone granules contain the following proportions, by weight, of the three ingredients mentioned:

| | Parts |
|--|-------|
| Powdered fused quartz ----- | 60 |
| Powdered silica gel ----- | 42 |
| Colloidal silica (18 percent suspension in water)----- | 8 |

In such a mixture, the ratio of fused quartz to silica gel is slightly over 1.4 to 1. When this ratio is increased, as

mentioned, the density of the spheres produced approaches 2.2 grams per cubic centimeter. As the ratio decreases, the silica gel predominates, and a low density, highly porous structure results.

For the spheres contemplated for possible use in telephone apparatus, the range of densities most usually desired is obtained when the composition of the siliceous mixture falls within the following limits:

| | Parts by weight |
|--|-----------------|
| Powdered fused quartz ----- | 50-70 |
| Powdered silica gel ----- | 30-50 |
| Colloidal silica (18 percent suspension in water)--- | 4-12 |

For other application, where a wider selection of densities in the fused silica spheres may be desired, compositions falling within the values given below are preferred:

| | Parts by weight |
|--|-----------------|
| Powdered fused quartz ----- | 40-80 |
| Powdered silica gel ----- | 20-60 |
| Colloidal silica (18 percent suspension in water)--- | 4-12 |

Though, as mentioned, the ratio of fused quartz to silica gel in the mixtures may vary between zero and infinity, from a practical viewpoint spheres whose densities encompass nearly all values of interest are obtainable when the compositions of the forming mixture lie within the ranges immediately following:

| | Parts by weight |
|--|-----------------|
| Powdered fused quartz ----- | 10-90 |
| Powdered silica gel ----- | 10-90 |
| Colloidal silica (18 percent suspension in water)--- | 4-12 |

As a very rough rule of thumb, the product density may be considered as a linear function of the percentage of fused quartz present in the mixture of fused quartz and silica gel. The empirical relationship

$$\rho = 0.5 + 0.017P$$

where ρ = density in grams per cubic centimeter and P = percent fused quartz in the fused quartz-silica gel mixture, will make it possible to gauge the approximate relative proportions of fused quartz and silica gel to be mixed for a desired density. Other factors, namely, the alkali metal ion content of the silica gel used and the extent to which the gel-containing mixtures are fired before spheroidizing, are also important factors in determining density, however, and may modify the composition estimated from the empirical expression given above. These other factors are considered below. Once standardized, the process will permit spheres whose densities are reproducible to within ± 0.3 grams per cubic centimeter to be manufactured.

For the total weight, 102 parts, of fused quartz and silica gel mentioned, 8 parts by weight of colloidal silica, in suspension, have been found to give sufficient binding action, independent of the relative amounts of fused quartz to silica gel present.

In the process of forming fused silica spheres, the silica gel and fused quartz are generally mixed as solids. These ingredients are to be finely ground to permit the intimate mixing required for homogeneity in the final product. Both the mentioned ingredients are preferably powdered to pass at least a 325 mesh sieve, or equivalent, having screen openings of 0.044 millimeter as specified by the United States Standard Screen Scale. Thorough mixing of the ingredients is conveniently accomplished by ball-milling the dry ingredients for about an hour.

The colloidal silica used as a binder is next added, conveniently as an aqueous suspension in water containing 18 percent of silica. Such a suspension has been available commercially under the trade name "Nalcoag," from the National Aluminate Corporation. Other suspensions, higher in silica content, may also be obtained and diluted to about 18 percent strength if desired.