United States Patent Office 3,523,028
Patented Aug. 4, 1970

3,523,028

PLURAL PHASE CERAMIC BODY

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No Drawing. Filed Oct. 27, 1964, Ser. No. 406,921
Int. Cl. C04b 35/00
U.S. Cl. 106—39 2 Claims

ABSTRACT OF THE DISCLOSURE

A fine-grained, dense polycrystalline ceramic body has grain regions and grain boundary regions. The grain regions consist essentially of two immisible phases. One of said phases consists essentially of a barium titanate-calcium zirconate solid solution, the other of said phases consists essentially of magnesium niobium oxide. An impurity concentration of a lower oxide of uranium is located in the grain boundary regions.

This invention relates to a sintered ceramic composition, and more particularly to a fine-grained dense ceramic body.

In the conventional method for making a ceramic body by the sintering process, finely divided powder is compressed and heated. As the temperature is raised, the process known as sintering takes place, whereby the pressed body acquires a higher density because of shrinkage, usually at a temperature considerably below the melting point. In the ideal case, this sintering is accomplished by the fact that the surface energy of the body is less than that of the individual grains; and this energy is released at the boundary betwen grains.

However, under ordinary circumstances, at the sintering temperature the phenomenon known as grain growth also occurs, whereby one grain grows at the expense of the other. Since the outlines of the compressed body are fixed at this time, the phenomenon of grain growth usually occurs accompanied by the occurrence of voids of substantial size, a phenomenon referred to as overfiring.

It is generally recognized in the art that those impurities in the material which have a low solid solubility in the grains tend to concentrate in the grain boundaries and promote this growth. For example, it appears that less than 300 parts per million of sodium will accelerate the grain growth of barium titanate by a factor of 50. Because of this phenomenon, it has been the practice of the prior art to employ ultrapure materials in order to get a fine-grained high density ceramic.

It is an object of this invention to provide a fine-grained dense ceramic body that is not subject to the disadvantages of the prior art.

It is another object of this invention to obtain a body of fine-grained high density ceramic material without requiring the ultrapurification steps of the prior art.

It is a further object of this invention to provide a fine-grained dense ceramic body in which the impurities with limited solid solubility in the ceramic may be tolerated, and indeed in some cases may be deliberately added.

In general, the objects of this invention are attained by the provision of a fine-grained dense polycrystalline ceramic body having at least two phases wherein the grains are surrounded by impurities that are insoluble in either of the phases. More particularly, my invention is based

on the fact that instead of having a single phase ceramic with attendant impurities, I provide a composition that consists of two phases. Under these circumstances, while the insoluble impurities are still concentrated in the grain boundaries of the grains constituting each phase, the grains are unable to grow at the expense of each other because of their mutual insolubility, and a fine-grained high density material results.

The following examples in which parts and percentages are by weight unless otherwise stated, illustrate the more detailed practice of the invention but are not to be construed as limitative.

EXAMPLE I

Fine-grained barium titanate is well known to have desirable electrical properties. However, when barium titanate is contaminated with even a trace of sodium as noted above, large crystals result. If, however, I slightly adjust the composition in accordance with this invention so that the ceramic constitution consists of two distinct and compatible phases, barium titanate and $BaTi_2O_5$, which are immiscible, the sodium concentrates in the grain boundaries, but no grain growth occurs. In one preferred example, I accomplish this objective by adding 1½ parts magnesium niobium oxide (85 parts Nb_2O_5–15 parts MgO) to 98½ parts of barium titanate-calcium zirconate solid solution (97 parts $BaTiO_3$–3 parts $CaZrO_3$) which result in the formation of the two phases. It will be understood that the specific materials employed are secondary to the attainment of two mutually insoluble phases so as to prevent grain growth.

Should either phase be capable of forming solid solutions with other materials then fine grains of these solid solutions can be sintered in the same way as above always provided there are two distinct phases. For example, barium titanate and sodium niobate form continuous series of solid solutions, thus both phases in the example cited above can have niobates in solid solution. To repeat, it is only the insoluble impurities which affect grain growth.

EXAMPLE II

To the barium titanate-calcium zirconate composition with magnesium niobium oxide additive of Example I, I add an insoluble impurity, for example a lower oxide of uranium. As noted above, this concentrates in the grain boundaries without promoting grain growth. At the same time the uranium oxide, although present in very small amounts, has a resistivity which is low compared to that of barium titanate. Thus if the ceramic made in accordance with Example II is made into an electric condenser by applying electrodes, on the application of voltage, the presence of the semiconductor uranium oxide at the grain boundaries insures a uniform distribution voltage between the electrodes and thus permits the dielectric to be operated under a higher field strength.

What is claimed is:

1. A fine-grained, dense, polycrystalline ceramic body consisting of grain regions and grain boundary regions said grain regions consisting essentially of two immiscible phases, one of said phases consisting essentially of a barium titanate-calcium zirconate solid solution, the other of said phases consisting essentially of magnesium niobium oxide, said barium titanate-calcium zirconate solid solution constituting a very large majority by weight of said ceramic, said magnesium niobium oxide constituting a very small minority by weight of said ceramic; and an

impurity concentration of a lower oxide of uranium located in said grain boundary regions.

2. The ceramic body of claim 1 wherein said barium titanate-calcium zirconate solid solution is formed from 97 parts by weight $BaTiO_3$ and 3 parts by weight $CaZrO_3$ and is present in 98½ parts by weight and said magnesium niobium oxide is formed from 85 parts by weight Nb_2O_5 and 15 parts by weight MgO and is present in 1½ parts by weight.

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U.S. Cl. X.R.

252—63.5, 520; 106—46