HIGH DIELECTRIC CONSTANT COMPOSITES OF METALLOPHTHALAOCYANINE OLIGOMER AND POLY (VINYLIDENE-TRIFLUOROETHYLENE) COPOLYMER

This application claims priority from U.S. Provisional Application No. 60/283,755, filed Apr. 13, 2001, and is a continuation-in-part application of U.S. application Ser. No. 10/108,231, now U.S. Pat. No. 6,787,238, filed Mar. 27, 2002, entitled "Terpolymer Systems for Electromechanical and Dielectric Applications".

The United States Government has certain license rights to the invention that is the subject of this application as a result of the development thereof under ONR Grant Nos. N00014-00-1-0623 and N00014-96-1-1173; under NIH Grant No. 1 RO1 HL65959; and under DARPA Contract No. N00173-99-C-2003.

BACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates to composite materials with elevated electric field induced strain levels, elevated dielectric constants, and having flexible mechanical properties. The materials of the invention comprise a polymer matrix and high dielectric constant organic material.

2. Description of the Prior Art

Because of their many attractive features such as light weight, high mechanical flexibility, and conformability, electroactive polymers are of great interest for a broad range of applications such as electromechanical and dielectric devices. On the other hand, compared with ceramic materials, the electroactive properties of polymers, such as the dielectric constant and electromechanical response, are not very high. In the past several decades, there has been a great deal of effort to develop polymers with improved functional properties. One of the focuses of these research and development efforts is to raise the dielectric constant of polymers substantially. In addition to applications directly related to capacitors and charge storage devices, a high dielectric constant polymer is also required for high electromechanical responses. The reason behind this is that in the electromechanical transformation process, the mechanical energy output of an electroactive material can't exceed the input electric energy due to the principle of energy conservation. The input electric energy is directly proportional to the dielectric constant of the material and also the square of the applied field. Therefore, in order to develop high performance electroactive polymers for electromechanical applications, one of the key issues is how to substantially raise the dielectric constant of the polymer.

Recently, by making use of high energy electron irradiation, it has been shown that the room temperature dielectric constant of poly(vinylidene-trifluoroethylene) copolymer P(VDF-TrFE) can be increased to about 50, a marked improvement compared with the current available polymers. Elevated dielectric constant PVDF-based terpolymer materials are also developed. However, how to raise the dielectric constant to much higher level without stiffening the polymers is still a challenge. For example, using high dielectric constant ceramics as the filler, the room temperature dielectric constant of the so-called 0–3 composites (ceramic powder polymer matrix composites) can reach 300. One of the consequences of this ceramic filling process to the 0–3 composites is to increase the elastic modulus of the composites significantly above that of the polymer matrix. It is an objective of the present invention to develop alternative 0–3 composite approaches to achieve high dielectric constant while maintain the high flexibility of the matrix. The present invention shows that by making use of the organic solids which possess high dielectric permittivity through the mechanism of nomadic polarization as the fillers, at room temperature and 100 Hz, a composite material with dielectric constant more 1,000 and dielectric loss less than 0.5 can be realized. The composite film is also mechanically flexible with the elastic modulus nearly the same as that of the polymer matrix. In addition, the composite films which elastic modulus is 0.6 GPa also exhibit high strain (~2%) under an electric field of 13 MV/m, a marked improvement compared with other electroactive polymers.

Accordingly, it is an object of the invention to provide polymer matrix materials that exhibit elevated room temperature dielectric constants.

It is another object of the invention to provide polymer matrix materials comprising high dielectric constant material.

It is yet another object of the invention to provide polymer matrix materials having desirable dielectric properties that have mechanical properties similar to their polymer matrix.

These and other objects and advantages of the present invention and equivalents thereof, are achieved by composites useful for electrical applications.

SUMMARY OF THE INVENTION

Using solution casting method, a high dielectric constant composite based on copper-phthalocyanine (CuPc) oligomer and poly(vinylidene fluoride-trifluoroethylene) copolymer is developed. The experimental data show that the low field dielectric constant of the composites (with 55 wt % CuPc) can reach 1,000 and the loss is about 0.5 at room temperature and 100 Hz. Due to the long range electron de-localization in CuPc which results in a strong space charge response of the composite to the external field, there is a strong frequency dispersion of the dielectric properties. In addition, the dielectric properties also exhibit a nonlinear behavior with electric field. One of the uniqueness of the composite is its mechanical properties which remain very much the same as those of the polymer matrix. Even for a composite with 55 wt % CuPc (the volume fraction of CuPc in the composite is also in the similar range), the composite film is still flexible with a Young's modulus of 1.2 GPa at room temperature. Furthermore, it has been demonstrated that in a composite with 40 wt % CuPc filler and relaxor P(VDF-TrFE) matrix, a strain of near 2% can be induced by a field of 13 MV/m while the composite modulus is 0.6 GPa. The strain is proportional to the square of the applied electric field. In addition to the simple composite approach, other approaches to achieve high dielectric constant with metallophthalocyanine oligomer are also discussed.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. **1** shows the molecular structure of copper-phthalocyanine (CuPc) oligomer.

FIG. **2***a* and FIG. **2***b* are graphs of the dielectric constant and dielectric loss, respectively, of composite films of the invention measured at room temperature. The films have different weight percentage of CuPc filler.

FIG. **3** is a graph of the dielectric properties of composites of the invention having 55 wt % CuPc measured at room temperature in the frequency range from 1 to 100 Hz.