

increased probability of electron tunneling between molecules, resulting in a higher dielectric constant and loss. After mixing with P(VDF-TrFE) matrix, the nomadic polarization of CuPc still plays an important role in the composite film, especially for the composite with high percentage of CuPc.

The composite prepared here also exhibits a high field induced strain, which is proportional to the square of the applied electric field  $E$ . Under a field of 10 kV/cm, a strain of 0.3% has been achieved. In the current composites with 55 wt % CuPc, the breakdown field is at about 10 kV/cm (1 MV/m). Therefore, by improving the breakdown field, this composite should exhibit a very high strain response under a relatively low field (10 MV/m, for example). Indeed, a 40 wt % CuPc composite which was electron irradiated (30 Mrads at 1000° C.) exhibits a field induced strain of near 2% under an applied electric field of 13 MV/m (FIG. 4) and the modulus of the composite is 0.6 GPa.

The temperature dependence of the low field dielectric constant and dielectric loss of the composite with 55 wt % CuPc was also investigated and the results are shown in FIG. 5. The data shows that over a relatively broad temperature range, the dielectric constant is quite high, especially at lower frequency, e.g., 100 Hz. A dielectric maximum of about 2,300 (at 100 Hz) was observed at 70° C., which is near the Curie temperature of P(VDF-TrFE) copolymer. For the non-irradiated copolymer, the dielectric constant is 17 at room temperature and 100 Hz and increase with temperature. At the ferroelectric-paraelectric phase transition temperature, which is about 70° C. for the copolymer studied in this article, the copolymer exhibits a dielectric maximum, which is about 50. Therefore the dielectric constant of the composite, which is determined by copolymer and CuPc, also show a dielectric maximum. This is consistent with the results presented in FIG. 5.

In addition to the metallophthalocyanine oligomer as the high dielectric constant filler, there are several other classes of high dielectric constant organic molecules, whose polarization is also based on delocalized electrons, which can also be used as filler: ClAn/Cl<sub>4</sub>Pa and Pyrene/o-iodoBA, whose molecular structures are illustrated in FIG. 6.

In addition to the P(VDF-TrFE) copolymer as the matrix, other copolymers with dielectric constant higher than 20 can also be used as the matrix of the composites such as the high energy irradiated P(VDF-TrFE) and P(VDF-TrFE) copolymers, the PVDF based terpolymers.

In addition to the metallophthalocyanine oligomer as the high dielectric constant filler, there are several other classes of high dielectric constant organic molecules, whose polarization is also based on delocalized electrons, which can also be used as the filler: ClAn/Cl<sub>4</sub>Pa, whose molecular structure is illustrated in FIG. 6a (ClAn/Cl<sub>4</sub>Pa,  $\delta_r=240,000$ ,  $\text{tg}\delta=0.831$ ), and Pyrene/o-iodoBA shown in FIG. 6b (Pyrene/o-iodoBA,  $\epsilon_r=4000$ ,  $\text{tg}\delta\sim 0.5$ ).

Additional approaches for high dielectric constant polymers based on metallophthalocyanine oligomer. In addition to the simple mixing approach as presented in the preceding sections, there are other approaches for dielectric constant polymers based preferably on metallophthalocyanine oligomer which can significantly reduce the loss and raise the breakdown field. In these approaches, CuPc will be directly incorporated into polymer chains at the molecular level. In one approach, CuPc oligomer is grafted with soft polymer chain such as polyethylene glycol (PEG). In CuPc molecule, there are 16 carboxyl groups (—COOH) attached to the CuPc conjugated ring. Those groups can react with hydroxyl group (—OH) on the PEG chain. The dielectric

property and mechanical property will depend on how many —COOH groups in CuPc are reacted with PEG and the molecular weight of PEG. For the case when the molecular weight of PEG is high enough, the grafted CuPc can be used directly as composite (liquid crystal polymer type).

CuPc can be directly grafted to PVDF-TrFE polymer chain. The advantage here is that PVDF-TrFE has a relatively high dielectric constant compared with other polymers. In this process, the reaction of PVDF-TrFE copolymer in aqueous NaOH with the use of phase transfer catalyst results in dehydrofluorination. As a consequence, —CH=CF— and CF=CF— double bond units will be formed. The copolymers with double bond units are treated further with peroxidate to form hydroxyl (—OH) group. PVDF-TrFE copolymer with —OH group is then grafted on CuPc by means of esterification.

Another approach addresses the issue of raising the breakdown strength of the composites in which the CuPc powder (or other high dielectric constant semiconductor organic molecular solids) is physically mixed with the polymer matrix. By working with small crystallites (for example, nano-size CuPc fillers) and preventing the formation of agglomerates, the effective field across the grain boundaries will be reduced dramatically which can improve the dielectric strength markedly. In addition to ultrasonic stirring to be used in the preparation of the composites to break up any possible agglomerates small amount of suitable surfactants can be added to the composites to improve the dispersion of CuPc (or other high dielectric constant semiconductor organic molecule solids) in the polymer matrix.

Another approach is a blocking layer approach. In this approach a blocking layer (such as one layer of P(VDF-TrFE) based electrostrictive polymer (the terpolymer or high-energy electron irradiated copolymer)) will be coated on the composite layer. The breakdown field of a thin layer of irradiated P(VDF-TrFE) polymer has been measured to be above 300 MV/m for a 20  $\mu\text{m}$  thick film. In this blocking layer approach, the PVDF layer thickness can be  $\sim 0.1 \mu\text{m}$ . Such a thin layer will have a much higher breakdown field since the breakdown field is inversely proportional to the film thickness due to the avalanche nature of the electric breakdown process. In addition, the thinness of this layer will not affect the dielectric constant of the whole composite very much. The effect of such a blocking layer is very much similar to the principle of the corona poling in which a field much higher than the breakdown field of the sample to be poled can be applied without causing breakdown because of the limit in the current available.

Although the present invention describes in detail certain embodiments, it is understood that variations and modifications exist known to those skilled in the art that are within the invention. Accordingly, the present invention is intended to encompass all such alternatives, modifications and variations that are within the scope of the invention as set forth in the following claims.

What is claimed is:

1. A composite material comprising:

a polymer matrix; and

at least one high-dielectric constant organic material having a dielectric constant higher than 1,000;

wherein said polymer matrix comprises a dielectric constant measured at room temperature of at least 20 when measured in the frequency range from about 1 to about 100 Hz.

2. The composite of claim 1, wherein said high dielectric constant organic material is a semiconductor.

3. The composite of claim 1, wherein said high dielectric constant material comprises at least one material selected