Generating High Dielectric Constant Blends from Lower Dielectric Constant Dipolar Polymers using Nanostructure Engineering

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Abstract: It is a great challenge in dielectric polymers to achieve a high dielectric constant while maintaining low dielectric loss and high operating temperatures. Here we report that by blending two glassy state dipolar polymers i.e., poly(arylene ether urea) (PEEU, $K = 4.7$) and an aromatic polythiourea (ArPTU, $K = 4.4$) to form a nanomixture, the resulting blend exhibits a very high dielectric constant, $K = 7.5$, while maintaining low dielectric loss ($< 1\%$). The experimental and computer simulation results demonstrate that blending these dissimilar dipolar polymers causes a slight increase in the interchain spacing of the blend in its glassy state, thus reducing the barriers

for reorientation of dipoles in the polymer chains along the applied electric field and generating a much higher dielectric response than the neat polymers.

1. Introduction

Dielectric materials store energy electrostatically through various polarization mechanisms and release it by depolarization. Dielectric capacitors are unparalleled in flexibility, adaptability, and efficiency for electrical energy storage, filtering, and power conditioning[1–8]. Thus, they have been used widely in a broad range of modern power electronic systems such as medical devices, hybrid electrical vehicles (HEVs), filters, switched-mode power supplies, and power weapon systems[1–7]. These applications require capacitors possessing high energy density, low loss, and high operating temperature. Compared to ceramics and electrolytic capacitors, polymer-based capacitors are attractive because of low manufacturing costs and low dielectric loss, high reliability due to high breakdown strength, and graceful failure with an open circuit[1]. To meet the demand of continued miniaturization and increased functionality of modern electrical and electronic devices and systems, the energy density of dielectric polymer capacitors must be improved. Since the energy density of a polymer dielectric capacitor is proportional to the dielectric constant K , it is a key performance parameter that polymers of interest should possess a high dielectric constant and low loss.

In this paper, we present a practical and widely applicable mechanism for enhancing the dielectric constant through nanostructure engineering of dipolar polymers. Specifically, we show that by blending two strongly polar polymers, e.g., a poly(arylene ether urea) (PEEU, $K = 4.7$) and an aromatic polythiourea (ArPTU), $K = 4.4$, the resulting mixture exhibits a very high dielectric constant, $K = 7.5$ while maintaining low dielectric loss ($< 1\%$)[9,10]. Both structure

analysis and computer simulation results indicate that the nano (molecular)-scale mixing of the two polymers causes a slight increase of the interchain spacing in the glassy blend, thus reducing the barriers for dipole reorientation along the applied electric field and generating a high dielectric response without compromising the dielectric loss. To our knowledge, this is the first report on a polymer with such a high dielectric constant of 7.5 and sufficiently low loss (below 1%) to be used in capacitors. The results demonstrated here, which can be applied to many existing dipolar polymers, pave the way for a very low cost approach for creating “new” dielectric materials from existing ones, but with dramatically improved dielectric response. The blending increases significantly the dielectric constant of dielectric polymer films compared to the best state-of-the-art dielectric in current use (biaxially oriented polypropylene, BOPP, $K \sim 2.2$), while also tolerating much greater operating temperature[1]. These advantages can enable many more uses of high power density capacitors in portable and automotive systems, aircraft control, and advanced weaponry.

2. Experimental section

2.1 Blends of PEEU and ArPTU:

All chemicals for synthesizing PEEU and ArPTU were purchased from Sigma-Aldrich. ArPTU was synthesized via microwave-assisted polycondensation of diphenylmethane-diamino (MDA) with thiourea in N-methyl-2-pyrrolidone (NMP) with p-toluenesulfonic acid (p-TSOH) as a catalyst[10]. After purification, ArPTU was isolated as yellow powder, which was used for film processing. PEEU was synthesized from *m*-phenylenedioxy dianiline and diphenyl carbonate by thermal polycondensation as shown in Scheme S1[9]. The mixture of the two

monomers was stirred at 150 °C in vacuum for 4 hours, and PEEU powder was obtained through purification with ethanol.

The blend solution was prepared by dissolving 1 wt.% of ArPTU and PEEU in DMF. The thin films were prepared by casting the solution onto silicon substrates precoated with 40 nm of platinum. The films were kept in a drying oven under vacuum at room temperature for 4 hours, followed by heating to 70 °C overnight, 110°C for 12 hours and then annealing at 180 °C for 1 day.

2.2 Characterization:

The dielectric data was obtained by using a HP 4294A Precision Impedance Analyzer and a Novocontrol GmbH Concept 40 broadband dielectric spectrometer. The grazing incidence X-ray scattering data were collected using a Panalytical X'Pert PRO MPD diffractometer. The wavelength of X-ray was 1.54 angstroms. Background scattering was subtracted using JADE analysis software and then the peak position was calculated for the individual polymers and blends. Atomic force microscopy (AFM) was performed using a Bruker Dimension AFM in tapping mode. Thermal gravimetric analysis (TGA) was carried out in N₂ at a heating rate of 10°C/min using a 2050 TGA from TA Instruments. Differential scanning calorimetry (DSC) was carried out using a TA instruments Q2000 to probe the thermal behavior. The data were taken at a scan rate of 10°C/min during heating. The negative heat flow represents endothermic (heat absorbed) direction.

3. Results and discussion

In dielectric polymers, a necessary condition for achieving high dielectric constant is that they contain dipoles in the polymer chains. Strong coupling among the dipoles can lead to high

dielectric constant, as have been observed in semi-crystalline polyvinylidene fluoride (PVDF)-based ferroelectric polymers ($K > 10$)[2,4,11–13]. However, the strong coupling among dipoles causes large hysteresis loss, not desirable for most polymer capacitor applications. On the other hand, it is well known that strongly dipolar polymers that are weakly coupled exhibit low dielectric loss, but also low dielectric constant at temperatures far below the glass transition temperature T_g , due to constraints of the glassy structure on the dipoles. In contrast, at temperatures above T_g , the reduced constraints on the dipoles, due to increased free volume, lead to a large increase in dielectric constant. For example, polyvinyl chloride (PVC), a simple polymer glass, exhibits a large increase in dielectric constant after undergoing its glass transition, from $K \sim 3$ below T_g to $K > 9$ above T_g [14,15]. The penalty is that the dielectric loss also becomes high at temperatures above T_g (loss $> 5\%$) due to cooperative segmental motions in the rubbery state, which have long relaxation times. If excess free volume can be introduced in strongly dipolar polymers at temperatures far below T_g , a relatively high dielectric constant may be achieved without the penalty of high dielectric loss.

Based on these considerations, we investigate a class of nanostructured dipolar disordered polymers, i.e., polymer blends. Blending two polymers may create partial mismatches between two dissimilar polymer chains, resulting in additional free volume, thus, reducing the constraints to dipole reorientation under applied field in the glassy state and raising the dielectric constant without causing high dielectric losses. An aromatic polythiourea (ArPTU) and a poly(ether ether urea) (PEEU), see Scheme 1 for the chemical structures, were chosen as the blend components. ArPTU and PEEU have dipole moments of 4.5 Debye and 4.89 Debye, respectively, leading to relatively high dielectric constants, $K = 4.4$ and $K = 4.7$ for the two polymers in the glassy state. The T_g 's of both polymers are above 200 °C. As presented in Fig. 1(a), the 1:1 blend (by weight)

of the two polymers exhibits remarkably high and reproducible dielectric constant ($K = 7.5$) while maintaining low loss ($<1\%$). The inset in Fig. 1(a) shows the dielectric constants of the individual polymers. To our knowledge, this is the first report of a polymer with such high dielectric constant and loss below 1%. Fig. 1(b) displays the dielectric properties vs. temperature (at 10 kHz), which shows that the blend exhibits a high dielectric constant and low loss up to 120 °C. In order to examine the possible effect of any sub- T_g transitions (β and γ relaxations) on the dielectric constant of the neat polymers and 1:1 blend [16], we carried out a broad band dielectric spectroscopy study. As presented in Fig. 1(b) and Fig. S1, there are no sub- T_g transitions down to -150°C over a broad range of frequencies. The TGA data in Fig. S2 shows no weight loss below 250°C, thus confirming the thermal stability up to 250°C, which is a very desirable feature for high temperature operation.

Blends with different PEEU/ArPTU ratios were also prepared and their dielectric properties at room temperature and 1 kHz are summarized in Fig. 1(c) and Table 1. A very large increase in the dielectric constant was also observed in blends with different ratios of the two polymers. The dielectric loss of these blends (above and below a 1:1 ratio) shows a slight increase ($> 1\%$) compared to those of the constituent polymers and the 1:1 PEEU:ArPTU blend. The increase in dielectric constant may be attributed to the increased free volume. However, that increase in free volume can also result in larger scale polymer chain motions, which increase the dielectric loss ($>1\%$).

Grazing incidence X-ray scattering of the ArPTU, PEEU, and blend with 1:1 PEEU:ArPTU ratio was carried out to probe structural changes and the data are presented in Fig. 2. As shown in many earlier studies, the broad X-ray peak around $2\theta = 18^\circ$ in Fig. 2 for the neat polymers arises from inter- chain segment scattering in the amorphous state. The scattering data in Fig. 2

reveal: (i) there is only one broad X-ray diffraction peak for the 1:1 blend; (ii) the broad X-ray peak for the blend is at ca. $2\theta = 17^\circ$, indicating that interchain spacing in the blend is more than 5 % larger than those of the individual polymers. The expanded interchain spacing in the blend enables easier dipole reorientation to the applied field and leads to a higher dielectric constant compared with those of the neat polymers while maintaining low dielectric loss. These results indicate that the reduced constraints achieved by molecular engineering of the dipolar polymers in the glassy phase can significantly increase the dielectric constant without compromising the dielectric loss [17]. The AFM image of the blend with 1:1 PEEU:ArPTU ratio is presented in Fig. 3, showing uniform mixing of the two polymers in the blend at the nanoscale. The DSC data of the blend (Fig. S3) does not show two glass transition steps till 250°C , suggesting single phase behavior, which is consistent with the AFM data.

To provide further insight into molecular and nano-scale mechanisms responsible for the observed enhancement of the dielectric response in the blends, we also carried out simulations of ArPTU, PEEU, and blends of ArPTU:PEEU in various blend compositions. In the calculations of the dielectric properties, we combine the results from both molecular dynamics (MD) and density functional theory (DFT) simulations to obtain the permittivity. The permittivity tensor is divided into two parts:

$$\varepsilon_{\alpha\beta}(total) = \varepsilon_{\alpha\beta}(ionic) + \varepsilon_{\alpha\beta}(electronic) \quad (1)$$

The ionic contribution is calculated from classical molecular dynamics simulations using the relation

$$\varepsilon_{\alpha\beta}(ionic) = \frac{4\pi}{\Omega k_B T} \mathbf{M}_{\alpha\beta}, \quad (2)$$

where Ω is the volume of simulation supercell, k_B is Boltzman constant, T is temperature. $M_{\alpha\beta}$ is the covariance of two dipole moment components α and β , where the dipole moment is calculated as a summation over the atomic charges multiplied by corresponding coordinates. More details can be found in Ref. [17–21]. Due to unknown atomic geometries of the polymers, both ordered and disordered trial structures are created and annealed in the simulations. In the annealing, structures are equilibrated at 600 K for 1 ns, cooled down at a rate of 25 K/100 ps to 300 K, and equilibrated at 300 K for 1 ns. *NPT* ensemble is used in the annealing and Nose-Hoover thermostat is employed for temperature and pressure control[22,23]. The atoms interact via the classical force field reaxFF as implemented in the LAMMPS package[24,25]. The simulation cells contain $\sim 1,000$ -6,000 atoms. To study the blends, supercells are elongated along the z-direction to match the cell dimensions of PEEU and ArPTU. Various mixing models are used and polymer chains are also randomly oriented in the xy-plane to increase the flexibility.

The electronic contribution is calculated by density functional perturbation theory (DFPT)[26]. We have demonstrated in previous work that the electronic permittivity tensor is not much affected by the packing of the polymer chains, thus smaller supercells are used to save computational resources[27]. The potential structures are re-optimized in DFT. We use the package QuantumEspresso[28]. Ultrasoft pseudopotentials with exchange-correlation functional PBE and empirical van der Waals correction are employed [29–31]. The energy cutoffs of the electron wave functions and electron charge density are 35 and 420 Rydbergs, respectively.

The calculated electronic permittivity of PEEU is 3.305 and that of ArPTU is 3.658. Since these are quite similar, we linearly interpolate these results for the blends, which would require much larger supercells in DFPT simulations. When the electronic permittivity results are added to the ionic ones, a substantial enhancement is observed, which is consistent with the

experimental results. The blend simulations reveal a significantly larger specific volume (see Fig. 4(a)) due to an increase in interchain spacing, again in accordance with the experimental results. Comparison of simulation and experimental data of dielectric constant vs. blend composition (PEEU:ArPTU wt ratio) at room temperature is presented in Fig. 4(b). The calculated enhancements are smaller, probably due to the relatively small size of the simulation cell, which does not fully capture the effects of nanoscale morphology, inaccuracies in interatomic potentials, and relatively short simulation time. Fig. 4(c) and 4(d) present the side view and top view of the simulation unit cells, comparing the molecular structures of the neat polymers with blends, illustrating the mismatches existing between the chains in the blend simulation cell. As shown by the expanded views of blend molecular structures in Fig. 4(e), the mismatch of the polymer unit length and dipolar functional group locations cause complex inter-chain forces, resulting in the bending of the polymer chains of the blend. These stacked curved chain structures, on average, slightly increase the free volumes, i.e., sub-nanoscale voids, between the polymer chains, and consequently enhance the dielectric constant.

4. Conclusion

In conclusion, we demonstrate a low-cost approach to achieve dramatically higher dielectric constants while preserving the low loss and high operating temperature in strongly dipolar polymer systems. Specifically, we show that nanostructure engineering through blending of two dissimilar strongly dipolar polymers creates sub-nanoscale unoccupied volume, leading to polymers with a dielectric constant of 7.5 and a loss less than 1%. This advance paves the ways to generate high dielectric constant polymers from existing lower dielectric constant dipolar polymers.

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Table 1. Summary of the dielectric properties of the neat polymers and blends at 25 °C

Polymer	Dielectric Constant (1 kHz)	Loss (1 kHz)
PEEU	4.7	1.1%
ArPTU	4.4	0.64%
Blend (1:1)	7.5	0.77%
Blend (1:2)	7.9	1.62%
Blend (1:3)	8.6	1.84%
Blend (2:1)	8.3	1.45%
Blend (3:1)	7.4	1.35%

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Figure Captions:

Scheme 1. Schematic of chemical structures of aromatic polythiourea (ArPTU) and poly(arylene ether urea) (PEEU) used to form a blend.

Fig. 1. Dielectric data of the 1:1 blend of PEEU and ArPTU (a) as a function of frequency at room temperature, including the inset which shows the dielectric data of PEEU and ArPTU; (b) as a function of temperature at different frequencies. (c) Dielectric constant vs. blend composition (weight ratio of PEEU:ArPTU) at room temperature and 1 kHz. Data points are shown and the dashed line is drawn to guide the eye.

Fig. 2. (a) X-ray diffraction data of ArPTU and PEEU, and their 1:1 blend. Background subtracted data of (b) PEEU with peak at 18.6° , (c) ArPTU with peak at 18.6° and (d) blend data with peak at 17° . Wavelength of X-ray used was 1.54 angstroms.

Fig. 3. AFM images: (a) amplitude and (b) phase for the PEEU:ArPTU 1:1 blend, showing uniform mixing of the two polymers in the blend, consistent with the DSC data of Fig. S3.

Fig. 4. (a) Computational results of dielectric constant vs. specific volume for PEEU, ArPTU, and blends for various supercells with different PEEU:ArPTU weight ratios. (b) Comparison of simulation and experimental data of dielectric constant vs. blend composition (PEEU:ArPTU

weight ratio) at room temperature. Data points are shown and the dashed line is drawn to guide the eye. (c) From top to bottom, side view of the simulation unit cells of ArPTU, blend with 1:1 PEEU:ArPTU ratio and PEEU respectively. (d) From left to right, top view of the simulation unit cells of ArPTU, blend with 1:1 PEEU:ArPTU ratio and PEEU. (e) Expanded view of the molecular structure of the blend.

Aromatic Polythiourea

poly(arylene ether urea) (PEEU)

Scheme 1.

Fig. 1.

Fig. 2.

Fig. 3.

Fig. 4.

Supplementary Information

Fig. S1. Dielectric data of (a) ArPTU, (b) PEEU as functions of temperature measured over a range of frequencies.

Fig. S2. TGA data of 1:1 PEEU:ArPTU blend

Fig. S3. DSC data of 1:1 PEEU:ArPTU blend

Vitae

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