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## Progress in Polymer Science

Volume 156, September 2024, 101870

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# Polymer nanocomposites: Interfacial properties and capacitive energy storage

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<https://doi.org/10.1016/j.progpolymsci.2024.101870> 

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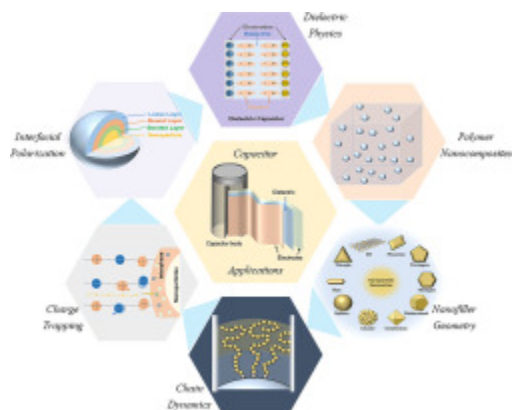
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### Abstract

An in-depth review is presented on the interfacial phenomena of polymer nanocomposites and the role of the interface/interphase in capacitive energy storage. The interaction between polymer chains and nanofillers upon filler dispersion and glass transition temperature are discussed through the lens of the adsorbed layer or polymer-grafted nanoparticles. Moreover, fundamentals of dielectric physics are discussed regarding charge transport and charge entrapment

on the interface, yielding the phenomenon of interfacial polarization. Therefore, the aim of this review is to inform the readers on the importance of the interface and highlight that both polymer chain dynamics and charge transport points of view are pivotal in the understanding of modern polymer nanodielectrics.

## Graphical abstract



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## Introduction

Polymers are an integral part of modern society with applications spanning every human activity from everyday life to advanced products applied in the microelectronics industry, biomedicine, and space exploration. The global plastic market size was estimated at US \$624.8 billion in 2023 and is expected to grow at a compound annual growth rate of over 4 % until 2030 [1]. On top of that, and especially now due to challenges arising from the climate crisis and the ever-increasing need for sustainable energy storage, innovations towards the use of new materials in energy storage applications are required. From 2017 to 2022, requirements for critical minerals to support the battery industry yielded a 300 % increase in the demand for lithium, and an increase of 70 % and 40 % in the demand for cobalt and nickel, respectively [2]. Therefore, the processability

and flexibility of polymers compared to other materials have renewed the scientific and industrial interest in the electrical/dielectric properties of polymers for energy storage applications. The addition of fillers of varying electrical characteristics lead to the formation of polymer composites that exhibit enhanced physical and chemical properties compared to neat polymers. When the fillers exhibit one or more spatial dimensions in the nanoscale, polymer nanocomposites or nanodielectrics are created, the properties of which strongly depend on interfacial characteristics.

The main takeaway from this review is the collaborative view employing both chain dynamics and dielectric physics towards a complete description of interfacial properties in polymer nanodielectrics. To achieve this result, the physical and chemical aspects of both the polymer matrix and the nanofiller need to be identified independently. While comprehensive in their coverage of various interface design methods and the impact of different filler types and morphologies, other reviews in this area do not delve as deeply into the fundamental physics governing these interactions. Instead of remaining at a surface-level discussion of interface design, this review explored the intricate details of how polymer chain dynamics and charge transport at the interface affect the performance of polymer nanocomposites. By examining the interaction between polymer chains and nanofillers through the lens of adsorbed layers or polymer-grafted nanoparticles, this review offered a more detailed analysis of the fundamentals of dielectric physics, including charge transport and interfacial polarization. Moreover, at the vicinity of the filler' surface, the polymer chains are governed by altered local chain dynamics that affect both the segmental relaxation and the charge carrier transport and entrapment. Therefore, the underlying fundamental mechanisms from both views are pivotal to understanding the interface/interphase and its role toward the capacitive energy storage response of polymer nanodielectrics. Therefore, by addressing these underlying mechanisms, the review not only provides critical insights that are necessary for developing high-performance polymer nanocomposites with optimized energy storage capabilities but also fills a significant gap in the literature by offering a comprehensive understanding of the interplay between polymer chain dynamics and charge transport at the interface.

The increasing interest of the research community in the fields of 'polymer capacitors' and 'polymer dielectrics' over the last 30 years is presented in Fig. 1a and 1b, respectively. It is evident that over the course of the last 3 decades, the US and Japan are continuously in the top 5 countries with the highest output of publications related to polymer capacitors and polymer dielectrics. However, the last 15 years have witnessed the emergence of new technological giants like India and China, with the latter being presently No.1 in the world in both areas. The intensification of research quantified by the increasing number

of publications highlights the need for a better fundamental understanding of physical concepts in the applied sector of polymer dielectrics for capacitor applications.

Here, a brief introduction of the three most important energy storage devices, namely batteries, supercapacitors and capacitors, is provided, with a focus on the latter and the advantages and challenges from the use of polymers and polymer-based composites. In Section 2, strategies to hinder the formation of aggregates by means of chain adsorption or polymer-grafting and the importance of nanofiller geometry are thoroughly discussed. Section 3 shows the effect that fillers have on polymers' molecular characteristics and the chain re-arrangement that affects structure and properties. In Section 4, the fundamentals of charge carrier transport in heterogeneous dielectric materials and the formation of a distribution of entrapped charges at the vicinity of the nanofillers' surface are discussed through the lenses of interfacial polarization. Finally, an overview of the present review is provided in Section 5.

The societal and economic challenges posed by climate crisis have shifted the energy sector towards the utilization of renewable sources of power to restrict the use of fossil fuels [3,4]. However, there are also technological obstacles stemming from the fact that targeted sustainable sources of energy generate power in a non-continuous fashion, for example, solar and wind energy parks that heavily rely on weather conditions [5]. Therefore, by efficiently converting energy into different forms (e.g., from thermal to electrochemical or electrostatic and vice versa), devices can store energy and subsequently supply it to the electrical grid reliably [6]. The principles of materials science and engineering are employed in the development of three major types of electrical storage devices, namely batteries, supercapacitors, and dielectric (electrostatic) capacitors. In the following part, the basic characteristics of these electrical storage devices are discussed.

A battery consists of two current collectors, connected to porous electrodes that facilitate the intercalation of ions. To facilitate ion transport, a battery requires the presence of an electrolyte (typically liquid) and an electronically insulating but ionically permeable membrane that acts as a separator. Energy storage and recovery are achieved by electrochemical reactions from the intercalation of ions between the porous electrodes (anode/cathode) and thus ionic mobility through the battery is required. As a consequence of ionic movements, the electrodes' polarities change, thus electrons flow through an external circuit. Typically, batteries are categorized as primary and secondary, i.e., devices that can be discharged gradually over their lifetime or charged/discharged repeatedly over hundreds or thousands of times. Optimization in the individual

components of a battery (electrodes, electrolyte, separator) can yield devices that are operational at various energy, power, and voltage ranges [[7], [8], [9], [10], [11], [12]].

Supercapacitors (also termed as “electrochemical capacitors”) resemble more a battery than a capacitor in terms of the mobility of charge carriers and device constituents. The phenomenon of electric double-layer is the fundamental underlying mechanism of supercapacitors that produces enormous capacitances and fast charge/discharge rates. The latter permits devices that are powered by supercapacitors to be charged in seconds, significantly faster than batteries that typically require hours [13]. Moreover, in applications where miniaturization of the energy storage device is required, thin film batteries often suffer from limitations like short cycle life and complex architecture that hinder their applicability [14]. By contrast, supercapacitors can be developed in devices with masses that range up to 2 g and can power small portable devices like MP3 players with very long cycle lives exceeding  $10^5$  cycles [13]. According to the Ragone plot presented in Fig. 2, supercapacitors can yield higher power than batteries and higher energy density than dielectric capacitors, thus placing them in between these two energy storage technologies [15,16].

The first steps toward the development of the dielectric capacitor was made by Ewald Georg von Kleist who invented the Leyden jar over 250 years ago [17]. Since then, the pursuit for better-performing dielectric materials for capacitor applications has been a continuous process, combining elements from electrical engineering, physics, chemistry, and materials science. Contrary to conducting or semiconducting materials (above their activation temperature), in ideal dielectric materials there is no macroscopic migration of charges, which is attributed to their high electrical resistivity. Therefore, insulators and high energy gap semiconductors ( $E_A > 3$  eV) are classified as “dielectric materials” or “dielectrics” [18].

A dielectric capacitor consists of an insulator sandwiched between two current collectors, i.e. electrodes. When an external electric field is applied, polarization phenomena occur as a result of induced or oriented dipoles. This happens because of induced dipoles, i.e., the stationary positive and negative charges at the electronic or atomic level are separated, or the re-orientation of permanent dipoles, which are randomly oriented in the absence of an external electric field [18,19]. Such dipoles are most often of molecular nature and relate to the chemical structure of the dielectric material under investigation [20]. As a result of the aforementioned polarization, an internal electric field is induced that is opposite to the external. The reorientation of dipoles produces dielectric dispersion and absorption phenomena that can be examined in the framework of

the dielectric relaxation that obeys the linear response theory for small electric field strengths [19,20]. As the dielectric material polarizes, charge carriers concentrate at the dielectric/electrode interface to cancel the polarization, leading to the charging of the capacitor and capacitive energy storage. Following the removal of the electric field, the dipoles' equilibrium state changes since there is no Coulomb force applied from an electric field, diminishing the polarization effects. This results in the release of charge carriers from the dielectric/electrode interface, and therefore the recovery of energy via the discharging of the capacitor [21]. Within the dielectric material, the movement of free charge carriers is considered a parasitic effect that weakens the energy storage (leakage currents) and as such, the formation of traps that capture free charges is encouraged [22]. The free mean path of such charge carriers inside a dielectric matrix is limited, indicating they travel short distances leading to low electrical conductivity values [23].

Dielectric capacitor devices find application in many modern electronic systems, namely in pulsed power systems used in hybrid electric vehicles or military applications, as small energy storage devices used in pacemakers, or in inverters for grid-connected green energy sources [24,25]. Able to balance the instantaneous power difference between the input source and output load, dielectric capacitors are employed in power converters due to their ability to discharge capacitive energy in a short period [26,27]. Power inverters occupy about 20 % of the mass of hybrid cars and require dielectric capacitors that can operate at temperatures exceeding 140 °C, a non-trivial task due to the presence of thermally activated charge carriers that increase leakage currents [[28], [29], [30], [31]]. Finally, at the forefront of renewable energy production, the efficient storage and recovery of energy is of particular importance, particularly for solar and wind energy sources that produce non-continuous power due to changing environmental conditions [6].

To be considered as a dielectric for high-energy capacitor applications, a material needs a specific combination of physical properties [28,[32], [33], [34], [35]]: (i) a high value of dielectric permittivity, which is a measure of the polarizability and therefore its ability to store energy; (ii) low dielectric loss ( $\tan\delta$ ) values, which translates to low intrinsic thermal losses associated with the alignment of dipoles under the influence of the external electric field and low electrical conductivity, which in return impedes leakage currents; and (iii) high dielectric breakdown strength so higher electrical fields can be physically endured by the dielectric material [[36], [37], [38], [39], [40]]. The generalized relationship that connects the value of the relative dielectric permittivity,  $\epsilon_r$ , and the intensity of the applied electric field,  $E$ , and their effect on the capacitive energy density ( $U_e$ ) is shown below at Eq. (1a) and Fig. 3a and specifically solved for a linear dielectric material at Eq. (1b)

and Fig. 3b:  $U_e = \int_0^{D_{max}} E dD U_e = \frac{1}{2} \epsilon_r \epsilon_0 E^2$  where  $D$  is the induced electric displacement following the applied electric field, and  $\epsilon_0$  is the dielectric constant of vacuum ( $8.854 \times 10^{-12}$  F/m). In linear dielectrics, the value of dielectric permittivity is independent of the applied electric field, with the latter being limited by only the dielectric breakdown strength  $E_b$ . Therefore, to develop dielectric materials with high energy storage capabilities, high values of both  $\epsilon_r$  and  $E_b$  are required [36].

At high electric fields, dielectric materials experience an increase in temperature as a result of Joule heating due to their resistive nature, indicating that the latter process prior to dielectric breakdown of the dielectric material is essentially a thermal event [42]. However, the electrical conductivity, being the product of the isothermal charge carriers' mobility and population, typically follows Arrhenius or modified Arrhenius temperature dependence [20,43], as it will be discussed later in Section 4.1 in detail. This is characteristic of hopping conductivity, which is the sudden displacement of a charge carrier to a nearby state that can be quantum-mechanically described with higher or lower energetic jumps of the potential barrier, with the latter being achieved through quantum tunneling [44,45], also discussed in Section 4.1.

To calculate the capacitive energy density of a dielectric material based on Eq. (1b), the maximum electric field at which the dielectric can be operational is squared indicating the importance of the dielectric breakdown field  $E_b$ . For this reason, polymer films are extensively used as dielectrics due to their strong insulating character and thus their high  $E_b$  values. For example, highly insulating polyolefins like cross-linked polyethylene (XLPE) and biaxially oriented polypropylene (BOPP) are widely employed as dielectric films in capacitor applications, with the latter being the industry's benchmark for polymeric membranes for the last 30 years [[46], [47], [48]]. However, polyolefins like polyethylene and polypropylene are non-polar materials and are therefore characterized by very low values of dielectric permittivity ( $\epsilon_r = 2 - 3$ ), thus negatively affecting the dielectric constant part of Eq. (1b) and decreasing the overall capacitive energy storage [48]. Moreover, BOPP suffers from low thermal stability that hinders its applicability at elevated temperatures (above 105 °C) which also strongly affects charge transport via the hopping of thermally-activated charge carriers [42]. To partially overcome this issue, highly polar, or high- $k$ , polymers are often employed, with room temperature dielectric constants exceeding 10. Typical examples include polyvinyl alcohol (PVA) and polyvinylidene difluoride (PVDF) and its copolymers and terpolymers. The dielectric constant of PVA is  $\sim 13$  at 1 kHz, which arises from the permanent dipole moment created by the polar hydroxyl groups within the polymer structure which strengthens the overall polarity of the material [49,50]. However, the high  $\tan\delta$  value of  $\sim 0.3$  at 1 kHz limits

its application in the energy storage field. It has been found that the dielectric constant of PVA can be increased by changing the crosslinking agents, which form the crosslinked cyanoethylated poly(vinyl alcohol) with a dielectric constant of 12.6 at 20 Hz [51]. As one of the most frequently used dielectric materials, PVDF exhibits a dielectric constant of  $\sim 10$  at 1 kHz due to the large dipole moment of the  $-\text{CH}_2-\text{CF}_2-$  repeating unit. Besides increasing the content of the highly polar  $\beta$  polymorph of PVDF through different processing conditions, the addition of bulky groups into PVDF chains is another way to enhance the dielectric constant of PVDF. The bulky groups can weaken the intermolecular interaction and promote an all-trans conformation to enhance polarity. Thus, the copolymer of poly(vinylidene fluoride-co-trifluoroethylene) (P(VDF-TrFE)) has a slightly higher dielectric constant ( $\sim 18$  at 1 kHz) [52]. The dielectric constants of poly(vinylidene fluoride-co-chlorotrifluoroethylene) (P(VDF-CTFE)) at 1 kHz is  $\sim 13$  [53,54]. Moreover, the terpolymers of PVDF usually exhibit a much higher dielectric constant due to the increased interchain distance, which facilitates friction-free dipole rotation in the presence of the electric field. The dielectric constant of P(VDF-TrFE-CFE), P(VDF-TrFE-CTFE) and P(VDF-TrFE-HFP) are 55, 47, and 30, respectively, all at 1 kHz [55]. On the other hand, novel dielectric polymers with high dielectric constants have been developed through molecular structure design in recent years [56]. Unfortunately, these materials often suffer from lower dielectric breakdown strength values or high costs associated with their composition or manufacturing conditions due to high glass transition temperature ( $T_g$ ) or melting temperature ( $T_m$ ) values [57].

Despite the use of high-k polymers, their dielectric constant is not high enough to revolutionize dielectrics for capacitor applications. Therefore, efforts have been made to increase their low dielectric constants without compromising their high breakdown strength, which will enhance their capacitive energy storage capabilities. Chemical functionalization has also been proven useful to increase the performance of dielectric polymers. The introduction of small polar groups with high dipole moment to the main polymer chain can induce secondary relaxations with intense dielectric strength that will contribute significantly to the overall polarization and hence dielectric constant of the material [56]. However, orientational movements of dipoles as a result of the applied electric field produce dielectric losses that can increase intrinsic thermal losses, an effect that can be described by an enhancement of the  $\tan\delta$  values. The presence of such side groups can modify the conformational structure of the main polymer chains thus altering the crystalline phase [58]. Still, even after employing chemical modifications, polymers and organic materials typically have relatively low dielectric constants (below 30) for ultra-high energy density values. However, the dipole moments in non-organic insulators and semiconductors are significantly higher-yielding materials with considerably higher dielectric constants [34,59]. However, these inorganic



materials suffer from low dielectric breakdown strength values, attributed mainly to charge transfer, typically due to electronic or ionic conduction from intrinsic carriers or injected carriers from the electrodes [60,61]. Therefore, to utilize the high dielectric breakdown strength values of polymers and the high dielectric constants of inorganic materials, composite materials composed of a polymer matrix and ceramic (nano)fillers have been the focus of dielectric research over the past two decades [32,33,62]. By decreasing the size of the nanoparticles, the interfacial area increases significantly, as shown in Fig. 4 below.

One of the drawbacks of using ceramic/oxide nanofillers in such applications is their hydrophilicity; the unwanted physisorbed water molecules induce ionic charge carriers that promote dielectric breakdown and failure, indicating that in some cases the presence of nanofillers might be disadvantageous compared to the use of neat polymers [63]. In addition to the electrical/dielectric properties, a dielectric system used in capacitive energy storage requires high thermal conductivity values towards the dissipation of heat caused mainly by the Joule effect. This is not a trivial problem since high thermal conductivities are caused by the movement of electron-phonon interactions (polaron quasiparticles) in electrical conductors, where the electrical and thermal conductivities are coupled in accordance with the Wiedemann-Franz law [64]. Electrical insulators like polymers are mostly characterized by very low thermal conductivities, on the order of  $0.1$  to  $1 \text{ Wm}^{-1}\text{K}^{-1}$  [65,66], and thus thermally conductive nanofillers can benefit nanodielectric systems. However, the typical enhancement in thermal conductivity rarely exceeds 30 % and is achieved for heavily doped systems that exhibit percolative behavior that negatively affect the electrical/dielectric properties and most importantly reduce the breakdown strength, even when insulators are used as nanofillers [67]. There are recent developments on the use of moderately and highly oriented polyethylene that exhibit ‘metallic-like’ thermal conductivities whilst being an ultra-low loss dielectric material, making it a potentially good candidate for future capacitive energy storage applications [[68], [69], [70], [71]].

The addition of fillers in polymer matrices for dielectric purposes extends the composite materials’ theory of combining the electrical properties of the individual materials, resulting in polymer composites with optimized behavior [72]. Although the concept is correct in principle, the geometry of the polymer composites is limited by the size of the fillers [73], and the resulting dielectric properties are often impaired due to the development of surface defects [[22], [74], [75], [76]]. By reducing the size of the fillers from the microscale down to the nanoscale, the aforementioned issues are resolved and the development of ultra-thin polymer nanocomposites, also called “polymer nanodielectrics,” is now a reality [[77], [78], [79]].

The various types of nanofillers and their importance based on their geometrical characteristics will be discussed in Section 2.

The incorporation of nanofillers in a polymer matrix has been observed to benefit the dielectric properties of polymers by increasing the dielectric constant and/or the dielectric breakdown strength, a fact largely attributed to the role of the interface/interphase [36,80]. As discussed in the previous section and shown in Fig. 4, the incorporation of nanoparticles with decreasing dimensions increases the polymer/filler interfacial area by orders of magnitude. The large interfacial area promotes localized chemical and/or physical interactions between the polymer matrix and the nanofillers, resulting in dielectric materials with enhanced macroscopic dielectric properties [81]. In addition, these interactions between phases often alter the dynamics of the polymeric chains, including the segmental mobility and the motions of polar groups [82]. Therefore, the region around the nanofillers should be considered to possess volume (interphase) and not just an area (interface) [83]. For example, in Fig. 4, if the interphase is assumed to be a 10 nm-thick spherical cell around each nanoparticle and the volumetric concentration is constant, then the interphase volume would increase by a factor of 12 and 425 for nanoparticles sized 100 nm and 10 nm, respectively, in comparison to the one sized at 1000 nm diameter. As expected, this new phase, the “interphase”, is characterized by different properties than the individual polymer matrix or inorganic nanofiller. This will be discussed in more detail from the scope of polymer chain dynamics (Section 3b) and dielectric physics (Section 4b).

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## Section snippets

### Polymer – nanofiller interactions

The interactions between the nanofiller and the polymer matrix depend on the design of the system, which greatly influences the dispersion state of nanoparticles and the resultant properties of the polymer nanocomposite. For instance, in the simplest case of adding bare spherical nanoparticles to an athermal (i.e., Flory-Huggins interaction parameter,  $\chi$ , equals zero) polymer, the nanoparticles exhibit a strong tendency to aggregate due to van der Waals forces and depletion interactions [84,85]. ...

## Crystal polymorphism – The case of PVDF

The morphology of crystalline domains in semicrystalline polymers is strongly determined by nucleation and crystal growth as part of polymer crystallization. In the presence of a foreign surface, the crystallization kinetics as well as the crystalline structure, orientation, and polymorphism (often referred to as a phase) can be altered, thus affecting the physical and chemical polymer properties. When there are favorable molecular interactions between the substrate and the crystallizing ...

## Charge transport and trapping interfacial effects

The presence, mobility, and interactions of charge carriers in dielectric materials has been an ongoing area of investigation for condensed matter physics for the past century. In the following Section, the effects of charge transport and entrapment of charge carriers will be examined from a purely dielectric physics point of view. Both hopping conductivity due to the presence of nanofillers and the localization of charges at the interphase that gives rise to interfacial polarization, strongly ...

## Concluding remarks

The applicability of polymer nanocomposites with interesting electrical/dielectric properties in insulation and energy storage applications is an emerging sector for research and development, particularly in light of the ever-growing climate crisis. One of the major aspects of polymer nanocomposites is their dependence on the interfacial interactions between the polymer chains and the nanofiller, and how these affect the macroscopic physical properties. To identify and discern the physical and ...

## CRedit authorship contribution statement

**Stavros X. Drakopoulos:** Writing – review & editing, Writing – original draft, Visualization, Project administration, Conceptualization. **Jiaen Wu:** Writing – review & editing, Writing – original draft, Visualization. **Shawn M. Maguire:** Writing – review & editing, Writing – original draft, Visualization, Project administration. **Sneha Srinivasan:** Writing – review &

editing, Writing – original draft. **Katelyn Randazzo:** Writing – original draft. **Emily C. Davidson:** Funding acquisition, Writing – review ...

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. ...

## Acknowledgements

The authors acknowledge support by the National Science Foundation (NSF) Materials Research Science and Engineering Center Program through the Princeton Center for Complex Materials (PCCM) (DMR-2011750) and the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, Catalysis Science program, under award DE-SC0022303. J.W. and E.C.D. are grateful for funds provided by the Princeton Alliance for Collaborative Research and Innovation.

S.S. and R.D.P. are grateful to the ...

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