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Opinion

High-*k* Polymer Nanocomposite Materials for Technological Applications

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Featured Application: High-*k* polymer nanocomposites are an important category of materials that demonstrate unique design possibilities, and offer excellent advantages with tunable properties for technological applications.

Abstract: Understanding the properties of small molecules or monomers is decidedly important. The efforts of synthetic chemists and material engineers must be appreciated because of their knowledge of how utilize the properties of synthetic fragments in constructing long-chain macromolecules. Scientists active in this area of macromolecular science have shared their knowledge of catalysts, monomers and a variety of designed nanoparticles in synthetic techniques that create all sorts of nanocomposite polymer stuffs. Such materials are now an integral part of the contemporary world. Polymer nanocomposites with high dielectric constant (high-*k*) properties are widely applicable in the technological sectors including gate dielectrics, actuators, infrared detectors, tunable capacitors, electro optic devices, organic field-effect transistors (OFETs), and sensors. In this short colloquy, we provided an overview of a few remarkable high-*k* polymer nanocomposites of material science interest from recent decades.

Keywords: actuators; dielectric properties; high-*k* materials; nanoparticles; polymer nanocomposites

1. Background

The discovery of polymers has given a new dimension to the present era: this relatively young subdivision of chemistry has been the topic of great development both as a basic and applied science over last five decades [1–6]. Generally, polymers are best known for their insulating properties because of the covalent bonds between the saturated carbon atoms. Since the properties of polymers can be altered by incorporating additives such as nano-fillers, many polymer frameworks were tailored and conveniently attained polymers with conducting/semiconducting behaviors with tunable properties opened new specialized applications in electronics [7–9]. The polymer structures with high-*k* dielectric behaviors were developed to create new interfaces in technological fields. The structural tunability of polymers in micro/nano electronics to develop miniature modules is always a challenging theme, where the polymers can be utilized not only as insulators, but also as conductive interfaces with the optimal tuning of electrical, mechanical and dielectric properties [10–16].

For a general understanding, the scale of the *k* value is fixed on the dielectric constant of silicon dioxide (SiO_2). The relative dielectric constant of silicon dioxide is 3.9, and the materials which possess $k < 3.9$ are commonly termed as low-*k* materials and those whose $k > 3.9$ are categorized as high-*k* materials [17–19]. Embedding high-*k* inorganic/organic hybrid nanomaterials into the dielectric polymers results in dielectric polymer nanocomposites with superior dielectric properties and high-breakdown strengths/high-energy density for suitable electronic applications [20–27].

Silicon dioxide (SiO_2) has been broadly utilized as a gate oxide material in metal–oxide–semiconductor field-effect transistors (MOSFETs). In recent decades, the gate capacitance of the silicon dioxide gate dielectric was improved by minimizing the size and thickness of the dielectrics in order to enhance the device performances [28–30]. Various high-*k* materials are used by replacing SiO_2 to diminish the leakage current and boost the power consumption, which perceptibly increases the gate capacitance of MOSFETs.

The capacitance *C* of the parallel plate capacitor is given by Equation (1):

$$C = \frac{\kappa \varepsilon_0 A}{t} \tag{1}$$

where κ is the relative dielectric constant of the material used (= 3.9 for SiO_2), *A* is the area of the capacitor and *t* is the thickness of the capacitor oxide insulator [31,32].

Since the dielectric polymer possesses a high electrical breakdown strength and the magnitude of total energy storage density, it depends on both the values of κ and the applied electric field, as polymer-based capacitors have proven their advantages over ceramic and electrolytic capacitors [33–35].

It is very important to know two main electrical parameters, the dielectric constant (ε') and dissipation factor (ε''), for microelectronic polymer dielectrics. The dielectric constant of a material can be defined as the ratio of the absolute relative permittivity of the material to the electric permeability of free space (i.e., vacuum). The magnitude of ε' depends on the amount of mobile (polarizable) electrical charges and the degree of mobility of these charges in the material. The ε' is temperature dependent, because the charge mobility depends on the temperature, the polarization of the material requires a finite amount of time, and frequency of the applied electric field [36–38]. In addition, it influences the measured dielectric constant.

The signal propagation velocity is given by Equation (2):

$$V_p = \frac{C}{\sqrt{\varepsilon'}} \tag{2}$$

where V_p is the velocity of propagation and C is the speed of light.

In an alternating current (ac) field, the dielectric constant is represented as a complex quantity, ε^*, and is the combination of a real component (dielectric constant = ε'), and an imaginary component, called the dielectric loss (ε''). This complex dielectric permittivity can be defined by the following Equation (3):

$$\varepsilon^* = \varepsilon' - j\varepsilon'' \tag{3}$$

With an increase in ac frequency, the charged particle's inertia inclines to preclude the displacement of the particles from keeping in phase. This leads to a frictional damping mechanism [39] which creates the power loss, because work must be performed to overcome these damping forces [40,41].

2. High-*k* Dielectric Polymers

Compared to conventional rigid silicon technology, the inherent desirable properties of high-*k* polymer nanocomposite materials offer a new dimension to the field of flexible and stretchable electronics [42,43]. The main benefits of polymer-based capacitor devices are unique in design with flexibility and the ease of processing. Therefore, no critical dimensions are required to utilize the materials to produce moderate high-voltage operating electronic devices as non-planar and flexible substrates [44,45]. Moreover, the incorporation of high-*k* nanostructured hybrid materials to the dielectric polymer matrix can regulate the mechanical stiffness and tunes the electronic properties. Thus, we can notice flourishing research in developing inorganic/organic hybrid nanomaterials and this contributed to the significant growth in accomplishing high-*k* polymer nanocomposites for technological applications [46,47].

Typically, enhanced dielectric breakdown strength can be achieved by loading nanoparticles into dielectric polymers. Firstly, a compatible solvent is used to disperse the nanoparticles and later embedding into the dielectric polymer matrix. This solution-mixing technique is the best known method to synthesize high-*k* polymer nanocomposite materials with superior dielectric breakdown strength [48–50]. Considering the breakdown strength, some of the important polymers are listed in Table 1. Based on breakdown strength [51–53], polytetrafluoroethylene (PTFE) and polypropylene (PP) will be the best choice [54,55]. However, the compatibility with solvents and the ease of thin film processing are also key parameters, which will decide the final application of the polymer composites. Because of the ease in thin film fabrication, polyvinylidene fluoride (PVDF) composites are an ideal choice. We notice plentiful research reports on PVDF composites on mechanical and acoustic sensors, actuators, energy harvesting and nonvolatile memory applications, because of its piezo-, pyro- and ferro-electric properties [56–61].

Table 1. Breakdown strength (dielectric strength) of some selected polymers.

Polymers	Breakdown Strength (MV/m)
Polytetrafluoroethylene (PTFE)	600–700
Polypropylene (PP)	640
Polyvinylidene fluoride (PVDF)	590
Polyethylene terephthalate (PET)	570
Polycarbonate (PC)	528
SU-8	440
Polyvinyl chloride (PVC)	140–210

Since SU-8 structure blocks can be photo-definable, photo-patternable high-*k* dielectrics on it can result in embedded capacitor applications [62–64]. Furthermore, polyvinyl chloride (PVC) is a widely used thermoplastic polymer, due to its versatile nature with plasticizers and high-*k* dielectrics nanoparticles, it can be successfully used in consumer electronics [65–70]. Due to the increasing demand for flexible and soft smart devices, our research group fabricated soft vibrotactile actuators based on silicon dioxide nanoparticles embedded in plasticized PVC gels. The soft gels were used as a dielectric layer in the designed vibrotactile actuators. To maximize the elastic restoring force, a wave-shaped ePVC gel was designed. The design of the soft vibrotactile actuator is presented in Figure 1 [70]. The proposed soft vibrotactile actuator based on plasticized PVC/silicon dioxide nanoparticle composites showed broad amplitude variation in a wide frequency range and created a variety of haptic sensations to the users.

To improve the robustness, processability and breakdown strength of the polymer nanocomposites, it is also important to consider the polymerization techniques, where the grafting of polymer brushes on inorganic nanoparticles can certainly enhance the compatibility of polymer–inorganic hybrid nanoparticles with dielectric polymer matrix [71–73]. Ellingford et al. defined that even by the intrinsic tuning of poly(styrene–butadiene–styrene), with polar organic groups such as methyl thioglycolate, results in self-healing dielectric elastomers as new actuators materials. The step to achieve these materials was via a one-step thiol–ene "click" reaction followed by low-temperature UV curing. The reported materials exhibited improved relative dielectric permittivity to 11.4 at 10^3 Hz, with a low dielectric loss [74].

A synthetic strategy developed by Kang et al. by combining hard silicon segments in a soft dielectric matrix was recently reported, where 1,6-bis(trichlorosilyl)hexane (C_6) was used as organosilane cross-linking agent to functionalized carboxy terminal liquid reactive rubber, dicarboxy-terminated poly(acrylonitrile-co-butadiene) (CTBN). Figure 2 represents a sketch for the formation of elastomeric network dielectric film from CTBN and C_6. They reported self-organized C_6 aggregates acting as nanofillers in the dielectric matrix, and enhancing the dielectric strength by inhibiting electrical treeing growth [75]. A similar strategy was reported by Lee et al., where statistical copolymer poly(styrene-co-methyl methacrylate) was designed via a reversible addition–fragmentation chain transfer (RAFT) process. Since the RAFT process involves ionic liquids, they reported a superior ionic conductivity of the resulting polymeric gel, as well as the enhanced device performance in transistor gating experiments [76].

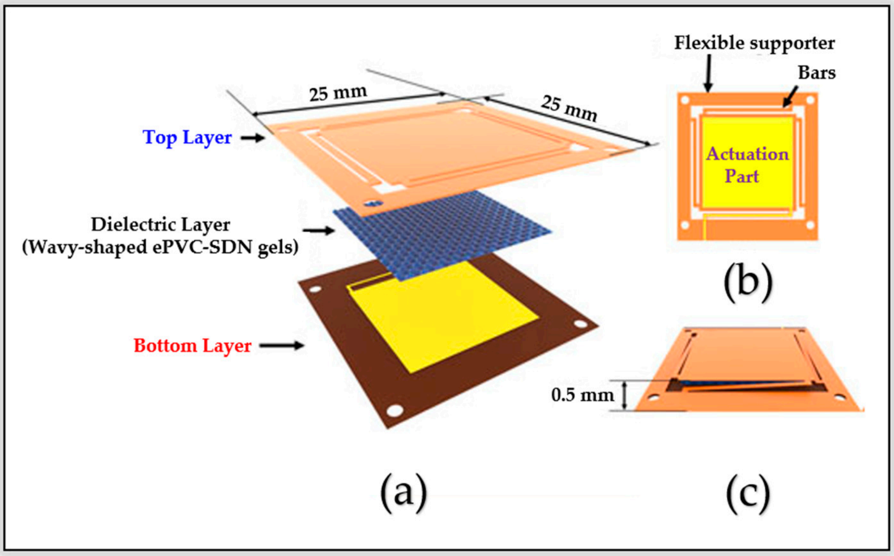

Figure 1. Illustrations representing the fabrication of the soft vibrotactile actuator based on ePVC–silicon dioxide nanoparticle (ePVC-SDN) gels. (**a**) Composition of the vibrotactile actuator. (**b**) Bottom surface of the upper layer. (**c**) Assembled vibrotactile actuator. Adapted with permission from [70], IEEE, 2018.

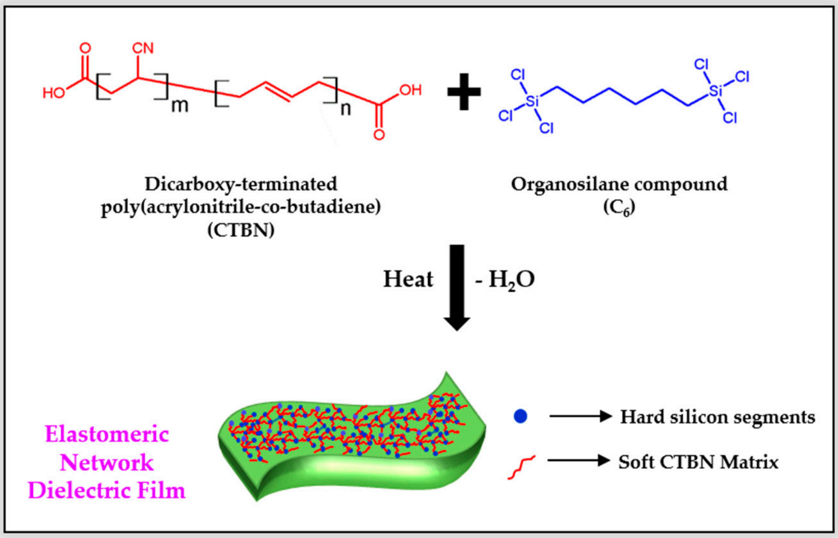

Figure 2. Schematic representation of the elastomeric network dielectric film formation using CTBN and C_6. Adapted with permission from [75], American Chemical Society, 2018.

The quasi-permanent dipole polarization or surface charge exhibited by polytetrafluoroethylene (PTFE) incited the researchers to use PTFE thin films produced by radio frequency (RF) magnetron sputtering or plasma-enhanced chemical vapor deposition [77]. Since PTFE possesses excellent chemical stability and dielectric properties, they are ideal to use as electret materials in organic electronics [78]. Murali et al. reported nearly isotropic and dimensionally stable silica-filled PTFE flexible laminates obtained by a hot pressing (SMECH process) technique for microwave circuit applications. The author reports the dielectric constant of 2.9 at the X-band frequency (8.2–12.4 GHz) for the maximum loading of fused silica (60 wt%) [78], whereas PTFE/rutile (rutile is a mineral primarily composed of titanium dioxide) nanocomposites, which exhibited a dielectric constant of above 7.0 at the X-band frequency for the 50 wt% loading of nano-rutile [79].

The polypropylene carbonate (PPC) dielectric film reported by Rullyani, et al. showed excellent compatibility with semiconducting pentacene and N,N′-Dioctyl-3,4,9,10 perylenedicarboximide (PTCDI-C8). Furthermore, the reported PPC film showed a surface energy of 47 mN m^{-1} with a dielectric constant of 3, which was utilized as a substrate material for organic thin film transistors (OTFTs) and organic inverters [80]. A sketch was drawn and presented in Figure 3, for understanding the basic design of OTFT and a bottom-gate top-contact OTFT on the PPC substrate. We can notice that an ultra-thin silver (Ag) metal gate was deposited on the PPC and thick layer (970 nm) of the biocompatible dielectric polyvinylpyrrolidone (PVPy) which was spin coated on the PPC layer. In addition, fabric-based wearable bioelectric and biochemical sensors were designed by loading silver nanoparticles in plastisols. These polymers can be easily screen-printed on textiles, since they adhere well to the fabrics [81]. It is evident that dielectric properties of polymer nanocomposites can be enhanced by the reinforcement of high-*k* dielectric nanoparticles, carbon allotropes, conducting polymers and organic crystalline materials [82,83]. However, polypropylene (PP)/carbon nanotube (CNT) nanocomposites reported by Zhang, et al. showed negative permittivity even at the low CNT loading, because of the low-resistance behavior of CNTs [84,85].

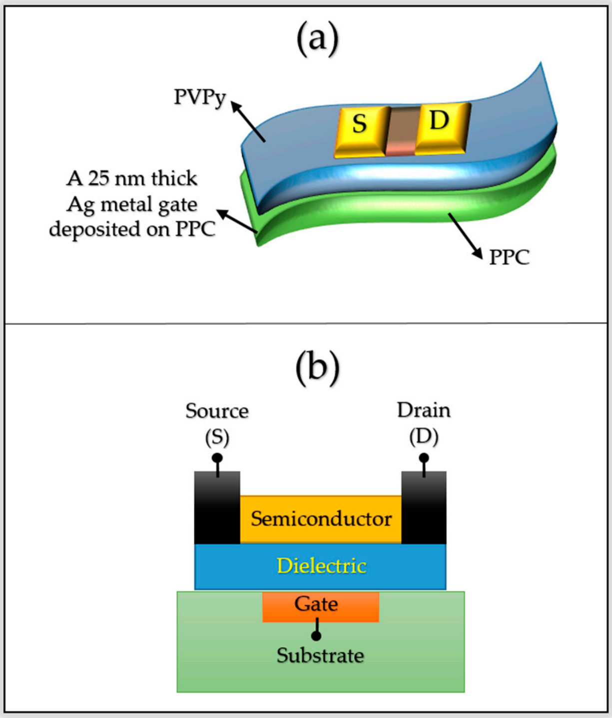

Figure 3. Pictograph representing (**a**) the bottom-gate top-contact organic thin film transistor (OTFT) on the polypropylene carbonate (PPC) substrate, and (**b**) a typical design of OTFTs. Adapted with permission from [80], Springer Nature, 2018.

Since poly(vinylidene fluoride) (PVDF) is a well known high-*k* polymer matrix showing the dielectric constant of about 12 at 1 kHz, a lot of research works were found. The flexibility, high dielectric permittivity, the piezoelectric, pyroelectric response, and the low acoustic impedance properties of PVDF demonstrate its potential applications in various electronic fields. A novel all-organic polyaniline–dodecylbenzenesulfonic acid (PANI–DBSA) and PVDF dielectric composites showed high-dielectric permittivity. For 20 wt% of PANI–DBSA doping to PVDF, resulted a dielectric permittivity of 150.0 at 25 °C [86], whereas PVDF and poly(vinylidene fluoride-trifluoroethylene) (PVDF-TrFE) filled with magnesium oxide nanofillers showed dielectric permittivity within the range of 10–22 at 25 °C [87]. Thomas, et al. reported composite thick films (thickness ≈ 85 μm) composed of PVDF/$CaCu_3Ti_4O_{12}$ nanocrystals with a relatively high dielectric permittivity of 90.0 at 100 Hz [88].

Today, polyester films or polyethylene terephthalate (PET) substrates have received considerable interest due to their inherent surface properties and designed engineering probabilities [89,90]. The PET substrates can be laying down to design thin film transistor arrays and in the construction of multimodal vibrational haptic interfaces [91]. Mi, et al. compared the properties of epoxy-coated (wood pulp) cellulose nanofibril (CNF) thin films with PVDF and investigated the microwave dielectric properties for potential broad applications in flexible high-speed electronics. They reported the dielectric constant of 2.6 for epoxy-coated-CNF and dielectric loss values in the range 0.03–0.042. However, the epoxy coated-CNF has proven to be more suitable for flexible microwave applications than for PET films [92]. Zhang, et al. reported that the coating of photoresist SU-8 on a silicon-(100) wafer substantially improves the flexibility and can be used for high-performance flexible electronics [93], whereas developed glass/SU8-gold electrodes by Matarèse, et al. were extremely transparent, and stable in the biological culture medium, which exhibited biocompatibility similar to glass [94]. Flexible and bendable (to 90°) tactile sensor arrays were also developed by Yeo, et al., consisting of aluminum nitride, based on micro-electro-mechanical system (MEMS) technology, where polydimethylsiloxane (PDMS) and a SU-8 photoresist layer were used as the supporting layers [95].

By the addition of various plasticizers, the mechanical stiffness and electrical permanence of PVC can be altered [96]. Even with the doping of modified inorganic nanoparticles, the fine-tuning of dielectric properties can be done and used to design high-performance actuators [70,97,98]. Some reports also demonstrate that the graphene oxide and plasticizer doping to PVC behaves as a soft actuator for artificial muscle applications [99–101]. The controlled robustness of plasticized thermoplastic PVC gels finds suitable applications in modular constructions of 3D-printable artificial muscles and sometimes the mechanical actuations are so effective they behave like human muscle [102–104]. More recently, ultra-high permittivity dielectric gels were fabricated and reported by Shi, et al. [105]. The fabricated dielectric gels were transparent, stretchable and showed a dielectric constant in the range of 30–50, offering great opportunities in soft robotics, sensors and optoelectronic applications [105]. The chemicals used and brief reaction conditions to obtain transparent dielectric gel reported by Shi, et al. [105] was sketched and presented in Figure 4.

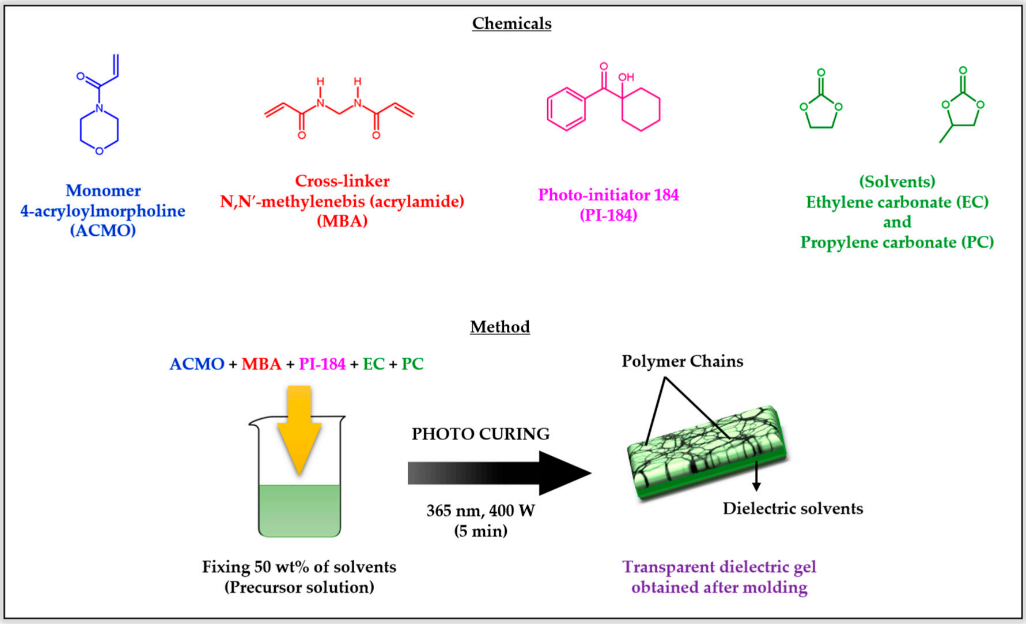

Figure 4. Schematic representation of the transparent dielectric gel formation using ACMO (monomer), MBA (cross-linker) and EC–PC solvents. Adapted with permission from [105], Springer Nature, 2018.

3. High-k Dielectric Nanoparticles

Many researchers have reported works on silicon dioxide nanoparticles in combination with other metal oxide nanoparticles and their polymer nanocomposites for sustainable energy storage and related applications [106,107]. In the construction of thin-film transistors (TFTs), zinc oxide (ZnO) nanoparticles are predominantly used. In the recent past, different kinds of oxide nanoparticles were explored, whereas perovskites show unique characteristics. Since then, lots of active studies have been done in constructing organic/inorganic structures to optimize the optical, dielectric, piezoelectric, electronic, catalytic or magnetic properties [108,109].

Nowadays, most studies have focused their attention on designing multi-metal–oxide hybrid nanoparticles because of their remarkable dielectric properties. Karmaoui, et al. prepared ultrafine strontium hafnium oxide ($SrHfO_3$) nanoparticles of 2.5 nm in size and demonstrated its potential as high-k gate dielectrics [110]. These perovskite-types, strontium-doped or mixed hafnium oxides with other metal oxides recently gained much interest because of exhibiting ferroelectric behavior and ultimately utilized in optoelectronic device applications [111–114]. The dielectric constant reported by Karmaoui, et al. was 17.0 and a relatively large capacitance value of 9.5 nF cm^{-2} [110]. Thus, these ultra-fine nanoparticles are readily useful in gate dielectrics for capacitors and in MOSFET technology [115,116]. However, hybrid CNTs decorated by ultrafine silver nanoparticles demonstrate the conducting behavior and show negative permittivity [116].

Dhaouadi, et al. studied the temperature-dependent dielectric behavior of nanostructured ferrite material, tetramanganese oxides (Mn_3O_4). The authors presented a convincing theory that the nano-dipole behavior of Mn_3O_4 under an applied electric field resulted in obtaining a high dielectric constant. This is due to the increasing dipole moment of nano-sized Mn_3O_4 particles per unit volume [117]. In addition, similar dielectric responses were observed in ZnO nanotubes and ZnO nanoparticles synthesized on a bio-template [118,119].

Some of the selected nanoparticles and its relative dielectric constants at ambient conditions are listed in Table 2.

Table 2. Dielectric constant of some of the selected nanoparticles.

Nanoparticles	Dielectric Constant	References
Cerium oxide (CeO_2)	4.1	[120]
$ZnMn_2O_4$	16.5	[121]
[a] Strontium hafnium oxide ($SrHfO_3$)	17.0	[110]
[b] Iron oxide (Fe_3O_4)	130.0	[122]
Cadmium sulfide (CdS)	163.0	[123]
$CoFe_{1.6}Al_{0.4}O_4$	200.0	[124]
$Ba_{0.9}Sr_{0.1}ZrO_3$	290.0	[125]
Carbon coated silver (Ag@C)	320.0	[126]
Cerium oxide (CeO_2)	370.0 [d]	[127]
$NiCr_2FeO_4$	900.0	[128]
$Pb(Zr_{0.97}Ti_{0.03})O_3$ coated silver	1700.0 [c]	[129]
$CaCu_3Ti_4O_{12}$	9000.0 [e]	[130]

[a] nanoparticle size = 2.5 nm, [b] stabilized by glucose, [c] measured at 200 kHz, [d] measured at 1 kHz, [e] measured at 100 Hz.

Currently, core–shell nanoparticle structures are gaining prominent attention due to their versatile architectures and wide applicability in electronic and optoelectronic devices [131]. Various materials have been synthesized with numerous nano architectures to understand the properties of organic polymers and its hybrid structures on loading inorganic nanoparticles [132]. Mahadevegowda, et al. investigated the aluminum (core)–aluminum oxide (shell) nanoparticles by coating nylon-6 polymer by a vacuum deposition technique. The fabricated core–shell nanostructures showed varied dielectric constants which were directly proportional to the thickness of the aluminum (Al) layer, with a relative permittivity of 28.0 reported for the 20 nm thickness of the Al layer [133]. By adopting a surface-coating approach in a solution followed by heat treatment, Hu, et al. synthesized a high dielectric constant titanium oxide-coated barium titanate (TiO_2@$BaTiO_3$) core–shell nanoparticle structures and embedded in the PVDF matrix [134]. The authors reported that the dielectric constant value obtained for neat PVDF was 9.2 and it was enhanced to 19.6 for 10-vol% of TiO_2@$BaTiO_3$/PVDF nanocomposites [134]. The amine functionalized carbon-coated Fe_3O_4/polyimide composite films showed a permittivity of 58.6 at 1 kHz, which for the Fe_3O_4@C–NH_2 composition was 1.13 vol% [135]. As reported by Ling, et al. we can notice exceptionally high permittivity values for the PVDF nanocomposites by loading novel titanium carbide@boehmite (TiC@AlOOH), and it was as high as 1.8×10^7 at 100 Hz when the content of the TiC@AlOOH nanoparticles was 41 wt% [136].

4. High-*k* Dielectric Nanocomposites

The dispersed conducting nanoparticles inside the insulating dielectric matrix phase can be explained by the percolation theory. The theory very well explains the variation of dielectric constant in the heterogeneous systems (see Figure 5). The dielectric constant values slowly increase and reach the maximum value at the percolation threshold (P_t). Where the conducting phase was separated by the optimal distance from the insulating dielectric phase, at the P_t point, the capacitive behavior of the nanocomposites can be noticed with excellent charge storage capability. The inhomogeneous distribution of the electric field inside the heterogeneous nanocomposites can dramatically increase the dielectric constant values [137]. Francis, et al. recently explored the high-*k* percolative nanocomposites based on multi-walled carbon nanotubes (MWCNT) and PVC [138]. The authors noticed a sharp change in the dielectric constant of the PVC nanocomposites, even with a small loading of MWCNT (4-wt%) to PVC. The heterogeneous MWCNT/PVC nanocomposite with 4% MWCNT concentration exhibited the dielectric constant of 13,066.

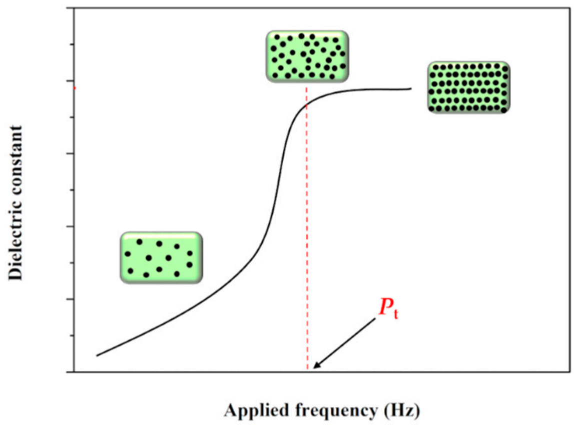

Figure 5. Sketch representing the percolation theory model, where P_t = percolation threshold.

Typically, homogenously dispersed ZnO nanoparticles in high-*k* resin (styrene-butadiene block copolymer, commercially known as K-Resin® KR20) were used for gate dielectric, with the aim of enhancing the dielectric permittivity [139]. Iacob, et al. testified the dielectric performances of raspberry-shaped iron oxide (Fe_3O_4) nanoparticles incorporated in PDMS, magnetite-rich PDMS nanocomposites showing a dielectric constant of 9.0 for a maximum loading of 60 wt% [140]. The authors specified the enhanced piezoelectric properties after embedding the iron oxide nanoparticles in the dielectric silicone matrix [140].

The dielectric constant values of some selected high-*k* nanocomposites are listed in Table 3.

Table 3. Dielectric constant of some selected nanocomposites.

Nano-Fillers	Dielectric Matrix	Weight % of Nano-Fillers	Dielectric Constant	References
Iron oxide (Fe_3O_4)	Polydimethylsiloxane	60.0	9.0	[140]
Copper/copper oxide (Cu/CuO)	Polypropylene	3.0	9.0	[141]
[a] Silicon dioxide	DGE-BA †	3.0	11.4	[142]
Aluminum oxide (Al_2O_3)	Polyvinyl alcohol	70.0	12.0	[143]
[b] $Ba_{0.2}Sr_{0.8}TiO_3$	Polyvinylidene fluoride	7.5	18.0	[144]
Silver and Nickel (Ag/Ni)	Polydimethylsiloxane	30.0	35.0 [c]	[145]
[d] $BaTiO_3$	Polyvinylidene fluoride	50.0	80.4	[146]
[b] $BaTiO_3$	P(VDF-TrFE-CFE) †	50.0	108.0	[147]
MWCNT/AgNP	Polyvinyl alcohol	1.0	620.0 [e]	[148]
Ni/$BaTiO_3$	Polyvinylidene fluoride	0.22 [f]	800.0	[149]
MWCNT	Polyvinyl chloride	4.0	13066.0	[138]

[a] nanoparticle size = 20 nm, [b] nanowires, [c] measured at 1 kHz, [d] modified by polyvinyl pyrrolidone (PVP), [e] measured at 100 Hz, [f] 0.22 volume fraction of Ni to $BaTiO_3$, † P(VDF-TrFE-CFE) = poly(vinylidene fluoride-trifluoroethylene chlorofluoroethylene), † DGE-BA = Diglycidyl ether Bisphenol-A.

Barium titanate ($BaTiO_3$), a well studied ferroelectric ceramic material exhibiting piezoelectric properties has broadly been used in energy storage and capacitor applications. From Table 3, we can determine that a high dielectric constant of 108.0 was achieved by loading nanowires of $BaTiO_3$ in P(VDF-TrFE-CFE). A loading of 50-wt% of surface modified $BaTiO_3$ by PVP to PVDF matrix can result in a > 80.0 dielectric constant. The strontium-doped $BaTiO_3$ shows comparably less dielectric constant (18.0) but with minimal (7.5-wt%) loading to PVDF. However, a very small volume fraction of Nickel to $BaTiO_3$ has drastically improved the dielectric constant of PVDF composites to a maximum of 800.0. We can also notice that with a minimum loading (3-wt%) of silicon dioxide to the DGE-BA polymer improved the dielectric constant to 11.4. Consequently, one should note the salient features of high-*k* nanoparticles, compositions of high-*k* dielectric polymeric matrix and other synthetic parameters, which open up a plethora of applications in organic electronics by tuning the dielectric properties.

With the advent of flexible electronics and advanced organic electronic power systems, practical applications for fabricating flexible polymeric dielectric nanocomposites became a more serious goal. The inclusion of multi-dimensional nano-fillers into dielectric polymers makes them desirable to use exclusively in the energy-storage applications [150–152]. Besides ceramic-based composites, the recent studies on polymer-based nanocomposites provide more advantage options for tuning desirable dielectric properties, low-temperature processability, mechanical flexibility with economically low-cost benefits. The acquired high-*k* material candidates find their active use in embedded capacitor applications [153,154]. Since the high dielectric constant and high-breakdown strengths of high-*k* polymer nanocomposites are the key factors to consider in designing various sensors and actuators, the incorporation of two-dimensional (2D) dielectric fillers such as boron nitride nano-sheets (BNNs) significantly tune the dielectric properties by improving the breakdown strength of high-*k* dielectric polymers [155–157]. The successful approach in this direction may emphasize its utilization in high-temperature-operating electronic vehicle applications [158].

5. Future Perspectives and Challenges

There is now an increasing tendency towards integrating technology and coherent classical routes to achieve high-*k* polymer nanocomposite materials. With this conditioned stimulus, the enhancement of the dielectric responses of high-*k* polymer nanocomposites with physiochemical and electromechanical stability is always a challenging motif. Since the stoichiometric aspect ratio and surface properties of nanoparticles to dielectric matrix also decides the ultimate properties for specialized applications, the development of functional organic moieties to alter the inorganic nano-architectures will need to be customized to acquire optimized high-*k* nanoparticles. A great deal of synthetic knowledge is also necessary, and this can bring more reliable high-*k* materials for organic electronics applications. Recent advances in designing soft actuators and electro-mechano responsive 3D-printable artificial muscles need engineering expertise and skills, which facilitate the comprehensive dynamic structures. Due to their outstanding capability of recoverable deformation, dielectric elastomeric materials have been explored to design sensitive smart materials for external stimuli. Whilst the properties of the dielectric elastomers were less researched, by embedding functionalized high-*k* dielectric nanoparticles.

The growing popularity of flexible electronics, also termed as flex circuits, is a technology to assemble electronic circuits on flexible/stretchable surface. This offers new solicitations in designing flexible and stretchable displays, flexible photovoltaic cell array panels, electronic circuits on fabrics, flexible wearable battery gadgets, etc. [159–162]. The incorporation of high-*k* dielectric nanomaterials into a variety of flexible polymeric materials can be seen as exactly the right strategy in designing new functional materials. Considering the key issue of low dielectric constant behavior of polymer dielectric materials, an effective fabrication approach is also a prerequisite to improve the dielectric properties and it has been an essential research topic in the development of high-performance high-*k* polymer dielectric materials.

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Conflicts of Interest: The authors declare no conflict of interest.

Abbreviations

BNNs	Boron nitride nano-sheets
CNF	Cellulose nanofibrils
CNTs	Carbon nanotubes
DGE-BA	Diglycidyl ether - Bisphenol-A
MOSFETs	Metal–oxide–semiconductor field-effect transistors
MWCNTs	Multi-walled carbon nanotubes
OFETs	Organic field-effect transistors
PC	Polycarbonate
PDMS	Polydimethylsiloxane
PET	Polyethylene terephthalate
PP	Polypropylene
PPC	Polypropylene carbonate
PTFE	Polytetrafluoroethylene
PVC	Polyvinyl chloride
PVA	Polyvinyl alcohol
P(VDF-TrFE-CFE)	Poly(vinylidene fluoride-trifluoroethylene chlorofluoroethylene)
PVDF	Polyvinylidene fluoride
PVPy	Polyvinylpyrrolidone
RAFT	Reversible addition–fragmentation chain transfer
RF	Radio frequency
SMECH	Sigma mixing, extrusion, calendering, followed by hot pressing
TFTs	Thin-film transistors

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