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B. GHULE, M. LAAD

Symbiosis Institute of Technology (SIT), Symbiosis International (Deemed University) (SIU)
(Lavale, Mulshi Taluka, Pune, Maharashtra 412115, India; e-mail: meena@sitpune.edu.in;
babaji.ghule.phd2019@sitpune.edu.in)

POLYMER COMPOSITES WITH IMPROVED DIELECTRIC PROPERTIES: A REVIEW

Materials exhibiting high dielectric constant (k) values find applications in capacitors, gate dielectrics, dielectric elastomers, energy storage device, while materials with low dielectric constant are required in electronic packaging and other such applications. Traditionally, high k value materials are associated with high dielectric losses, frequency-dependent dielectric behavior, and high loading of a filler. Materials with low k possess a low thermal conductivity. This creates the new challenges in the development of dielectric materials in both kinds of applications. Use of high dielectric constant filler materials increases the dielectric constant. In this study, the factors affecting the dielectric constant and the dielectric strength of polymer composites are explored. The present work aims to study the effect of various parameters affecting the dielectric properties of the materials. The factors selected in this study are the type of a polymer, type of a filler material used, size, shape, loading level and surface modification of a filler material, and method of preparation of the polymer composites. The study is focused on the dielectric enhancement of polymer nanocomposites used in the field of energy storage devices. The results show that the core-shell structured approach for high dielectric constant materials incorporated in a polymer matrix improves the dielectric constant of the polymer composite.

Keywords: polymer composites, dielectric constant, dielectric strength, ceramic filler, energy storage.

1. Introduction

Energy storage devices, bypass capacitors used in microelectronics, electronic devices working at high operating frequency widely use dielectric materials with high dielectric constants [1–3]. Miniaturization of electronic devices demands the low dielectric constant material for the packaging and insulating materials [4, 5]. Therefore, the synthesis of both types of materials has created a lot of interest among the researchers today. The materials with high dielectric constant should have high dielectric strength, low dielectric losses, chemical resistance, frequency-independent response, and good mechanical properties for its desired applications. It is hard to get the unique combination of all properties in one component material. Usually, ferroelectric ceramics show a high dielectric constant and low losses, but these materials are generally brittle in nature. Due to their

brittleness, it is difficult to fabricate a complex shape. Even the processing of ceramic substrates during the circuit fabrication [5] becomes difficult due to their brittle character. In addition, ceramic materials have poor dielectric strength. Polymers exhibit good dielectric strength, stability in chemical attack, hydrophobic and restraint in temperature, and are also easy to be treated, but have very small value of the dielectric constant ($k < 10$), which limits the scope of their application [1–3, 6, 7]. High dipole density polymers can have high energy density [8]. In addition, they have a high dielectric strength. Therefore, for high energy density capacitors, polymers are the best suited dielectric materials. To meet desired dielectric properties, composites of polymers with metals, semiconductors, metal oxides, and ceramics are synthesized using a variety of filler materials.

To achieve the desired properties in dielectric composites, the connectivity between filler and matrix materials is very important. In a multiphase mate-

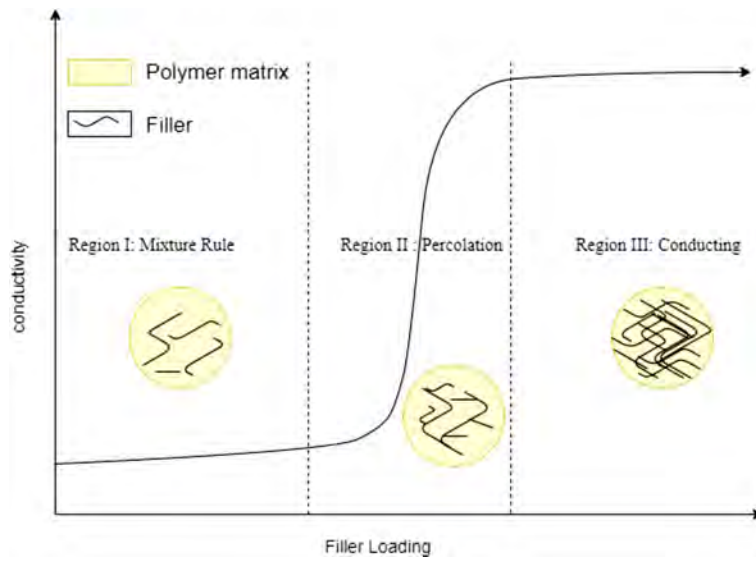


Fig. 1. Change in the conductivity of a filler in the matrix at various loading levels

rial, the interspatial relationships are described by the concept of connectivity which affects all properties of the composites. The filler dimension, interfacial properties, percolation threshold, and porosity affect the properties of composites [5, 9, 10]. Therefore, different factors are considered, while designing the composite material such as a matrix polymer, filler, size and shape of a filler, surface treatment of filler, hybridization of a filler, synthesis method, *etc.* [11]. This review article focuses on the thermoset, thermoplastic materials both polar and non-polar polymers like polyvinylidene fluoride (PVDF), polyvinylidene fluoride trifluoro-ethylene P(VDF-TrFe), cytoresin polymer, polypropylene (PP), polystyrene (PS) *etc.*

2. Theoretical Models for Composites

The relative permittivity (dielectric constant) of a material is related to its capacity of energy storage, when a potential is applied across it. The energy density stored in a material is related to the macroscopic property called the polarization [9]. The energy density (U) stored in a dielectric material is governed by the applied electric field (E) and electric displacement (D) as,

$$U = \int E dD. \quad (1)$$

For linear dielectrics,

$$U = \frac{1}{2} DE = \frac{1}{2} \epsilon_0 k E_b^2, \quad (2)$$

where, E_b , k , and ϵ_0 are the breakdown field strength, effective permittivity (dielectric constant) of the composite, and permittivity of vacuum, respectively. In nanocomposite materials, the loading level of a nanoparticle filler should be rationally chosen in such a way that the combination of k and E_b gives the maximum energy density [7, 8, 12–17]. In dielectric composites, especially those with the 0–3 connectivity, the percolation level plays an important role. The volume fraction of filler particles at which initially isolated inclusions get interconnected and distort the pure 0–3 connectivity called percolation threshold [18]. Particles of certain size and shape have clearance (matrix material) between neighbouring particles below the percolation threshold. The mean percolation threshold depends on the shape and size distributions of particles [9]. For conductive fillers, the percolation is defined as a change in the dispersion state of the conducting phase of a composite. The percolation threshold is the minimum volume content of the conducting filler, where the drastic change in the electrical conductivity begins [19]. Figure 1 shows changes in properties of polymer composites at the percolation threshold. It is clear from Fig. 1 that, as the volume fraction

of filler particles increases, initially isolated inclusions get interconnected with the polymer matrix and significantly affect the conducting properties of the matrix. Percolation theory of dielectric composite suggests,

$$k = k_0 \left| \frac{f_c - f_{\text{filler}}}{f_c} \right|^{-q}, \quad (3)$$

where, k is the effective dielectric constant of a composite, k_0 the dielectric constant of the matrix, f_c the percolation threshold, f_{filler} the volume fraction of a filler, and q is the critical exponent [20].

For the mechanical flexibility of PVDF, a low percolation threshold is required. It is obtained by introducing graphene nanoparticles in PVDF with large k ($k > 200$) at the percolation threshold f_c [21]. The research study reported on that, at a low percolation threshold, k up to 806 can be obtained by introducing SiC-CNT in PVDF [22].

The effective dielectric constant of a composite material is a function of the dielectric constant of individual constituents, filler loading level, size and shape of filler particles, dispersion of filler particles, dielectric displacement, *etc.* Various models are proposed to explain the dielectric properties of composites. Mixing equations proposed by theoretical models are based on electrostatic theories. Moreover, some mixing equations have a stronger empirical background. When the effective dielectric constant and volume fractions of individual constituents are measurable, then the properties of the material components can be derived using some mixing equations [9, 23].

Dielectric properties are also influenced by the dispersion of a filler in the matrix, morphology, and interactions between the two phases [5]. Many composites deviate from the ideal configuration due to the clustering of a filler and an increase in the porosity [23]. Theoretical models proposed to predict the dielectric constant of a composite are as follows:

1) Average Mixing Model

$$k = v_1 k_1 + v_2 k_2; \quad (4)$$

2) Maxwell-Garnett

$$k = k_2 + 2v_1 k_2 \left[\frac{k_1 - k_2}{k_1 + k_2 - v_1(k_1 - k_2)} \right]; \quad (5)$$

3) Lichtener formula (Classical model of logarithmic mixing rule)

$$\ln k = v_1 \ln k_1 + v_2 \ln k_2; \quad (6)$$

4) Power mixing rule

$$k^\beta = v_1 k_1^\beta + v_2 k_2^\beta. \quad (7)$$

By the Bitcher formula, $\beta = \frac{1}{2}$, and, by the Looyenga formula, $\beta = \frac{1}{3}$.

Here, k is the effective dielectric constant of composites, k_1 , k_2 , and v_1 , v_2 are the dielectric constants and the volumes of a filler and a matrix polymer, respectively [24]. Dielectric loss is a complicated phenomenon. Therefore, a relatively few theoretical models are available for the prediction of a loss factor or loss tangent. The general mixing model predicts theoretical values in good agreement with experimental data. We have

$$(\tan \delta_c)^\alpha = \sum v_{fi} (\tan \delta_i)^\alpha, \quad (8)$$

where $\tan \delta_c$, $\tan \delta_i$, and v_{fi} are the loss tangent of the composite material, loss tangent of i th material, and volume fraction of the i th material, and α is a constant which determines the mixing rule.

The relation $\alpha = -1$ means the serial mixing, $\alpha = +1$ the parallel mixing, and $\alpha = 0$ gives the logarithmic mixing rule [5]. It is affected by the presence of moisture or humidity. Dielectric losses in a material may arise due to intrinsic or extrinsic imperfections in the crystal lattice. The proper processing of materials minimizes the losses due to extrinsic imperfections. Dielectric losses due to different types of defects show different temperature and frequency dependences. A hydrophilic filler like Al adsorbing humidity can affect dielectric losses [5, 9]. Dielectric properties of composites are frequency-dependent. Different physical processes such as dielectric resonance, dielectric relaxation, electrical conduction, and loss from non-linear processes cause the dissipation of the electrical energy leading to dielectric losses in materials [9, 25].

Generally, the theoretical models applicable for low loading and increasing filler contents show diverse results. This occurs, because, at higher loadings, there is an imperfect dispersion of ceramic filler particles in the matrix and due to air trapped or the porosity of the composite. Another reason for this deviation

in results is due to the fact that a theoretical model does not consider the matrix–filler interphase interactions. These predictions are found to be in agreement for composites which have nearly same relative permittivity values for a filler material and the matrix [5, 9, 10].

3. Factors Affecting Dielectric Properties of Composites

3.1. Polymer material

Polymer composites show different properties, and those properties depend on whether the polymer used as a matrix is a thermoplastic polymer or thermoset polymer. Such property as the melting temperature is associated with a thermoplastic polymer, while the thermoset polymer shows a dimensional behavior which is characterized by its glass transition temperature (T_g). Thermoplastic polymer composites require no curing and show the unlimited shelf life, reprocess ability, lower moisture absorption, chemical inertness, higher service temperature, and high environmental tolerance compared to thermoset matrix composites, but show a relatively high viscosity and processing difficulties [26]. Even at a high filler loading (60 vol%), thermosetting composites exhibit a low porosity, but a high moisture absorption and low tensile strength. It is difficult to use thermoset composites in extreme chemical environments. Polyphenylene oxide (PPO), poly(tetrafluoroethylene) (PTFE), polyetherimide (PEI), *etc.*, are the major thermoplastic polymer matrices having frequency-independent dielectric properties in the microwave frequency region [9]. A research study has reported on that composites like $\text{Al}_2\text{O}_3/\text{PEI}$, MgO/PEI , SiO_2/PEI , $\text{BaTiO}_3/\text{PEI}$, BN/PEI polyetherimide (PEI) have dielectric constants independent of the nanofiller type [27]. Polymethyl methacrylate (PMMA) is good for low- k polymer composites [28].

The polar polymer increases the dielectric constant of a composite material at low frequencies, but has hardly any effect at microwave frequencies [5]. PVDF has low water absorption characteristics and, due to the polar nature, exhibits a high $k > 10$ value [8, 29]. Cynoresins have high polarity cyno groups as side chains which exhibit the dielectric behavior. Because of a polar backbone, the polar polymers show a strong affinity for inorganic oxides [30, 31]. It

was reported on the synthesis of low- k , low-density $\text{Mg}_2\text{SiO}_4/\text{PTFE}$ composite, with k value remaining almost constant throughout the frequency spectrum 0.5 to 3 MHz [23].

3.2. Filler materials

Research studies have reported on the use of a conductive or semiconductor filler which increases the interfacial polarization between a filler and the polymer matrix, thereby enhancing the dielectric constant of the composite [19, 32]. The conducting filler dispersed in an insulating matrix exhibits a high permittivity near the percolation point of the conducting phase [33]. A conductive or semiconductor filler leads to large dielectric losses. The use of highly polarizable conjugated polymers enhances the dielectric constant to a good extent, but the dielectric loss also increases proportionally, as the conduction occurs due to the formation of a conductive path [3, 20, 24]. Because of a conductive filler, the dielectric constant increases, but the dielectric strength significantly decreases [34]. For a conductive or semiconductor filler, the dielectric loss is large [9]. For a conductive filler, the dielectric constant of a composite depends on the interparticle distance. A variation in the critical value of the interparticle distance in composites is found to depend on the conductivity of the filler, nature of polymer used *etc.* [19]. With the help of a small-size ferroelectric ceramic filler, the high dielectric constant is obtained [1, 4, 35]. Ceramic nanofillers with large aspect ratio are preferred to their spherical counterparts for the improvement of E_b of nanocomposites [36]. The dielectric strength of SiO_2/PE composites does not depend on the degree of crystallinity [37]. K. Yang *et al.* reported dielectric properties for ZnO/PVDF composites with respect to the filler structure. For commercial ZnO , $k = 19.4$, and the dielectric strength is equal to 45 kV/mm; for flower-like ZnO , $k = 221.1$, and the dielectric strength is equal to 42 kV/mm; for walnut-like ZnO , $k = 104.9$, and the dielectric strength is equal to 40 kV/mm [32].

In PP/BaTiO_3 and PP/TiO_2 composites, metal oxide fillers do not affect the breakdown strength of a filler [16, 38]. Another group of researchers investigated transparent $\text{TiO}_2/\text{polyvinyl-alcohol(PVA)}$ and TiO_2/PMMA nanocomposites exhibiting high k values of 24.6 and 26.8, respectively [39]. The dielec-

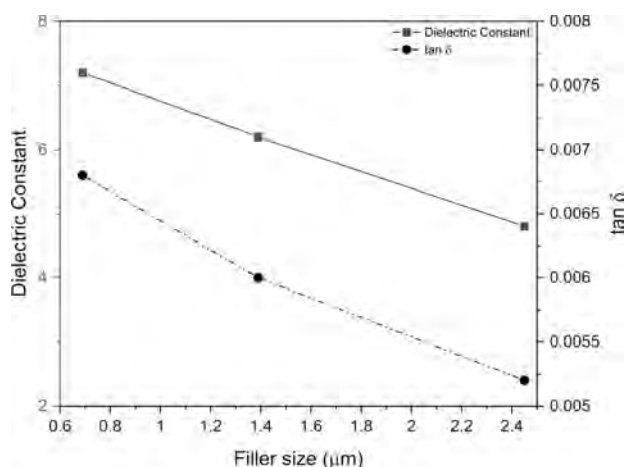


Fig. 2. Variation of the dielectric constant and dielectric losses with the filler size

tric constant of polydimethylsiloxane (PDMS) was reported to be improved by introducing graphene oxide as a filler by a research study [40].

3.3. Dielectric and thermally conducting fillers

The application of dielectric materials as fillers enhances the dielectric constant of a composite significantly. When a thermally conducting filler is used, the dielectric constant increases considerably, but the dielectric strength decreases drastically. The introduction of high dielectric constant materials like BaTiO₃, BST, BNNs enhances the dielectric constant, dielectric strength and gives a superior power density at very low volume [7, 12, 41]. S.H. Yao *et al.* showed that the addition of the thermally conducting filler Al₂O₃ decreased the dielectric strength of Al₂O₃/BaTiO₃/PP, while the addition of a high dielectric constant filler material increased the dielectric strength in Al₂O₃/BaTiO₃/PP [35].

3.4. Filler size and shape

The dielectric constant of a material mainly depends on the interfacial polarization. As the size of filler particles decreases, the effective surface area of a filler in contact with the matrix polymer increases leading to an increase in the interfacial polarization which further enhances the dielectric constant, but decreases the dielectric strength [6, 19]. N. Guo *et al.* synthesized nanocomposites and observed that rod-shaped nanofillers exhibited a higher dielectric con-

stant than the nanocomposite prepared using spherical nanofillers [16]. A research study has reported on that needle-shaped TiO₂ fillers increased the dielectric constant more than the spherically shaped TiO₂ fillers, when composed with polymeric cholesteric liquid crystals at a lower volume than spherical ones [42]. Another investigation reported on that, as the filler size was reduced to the nano scale, the dielectric loss increased effectively as nanoparticles exhibited a tendency to agglomerate. Nanocomposites possessed a higher dielectric loss than microcomposites, higher moisture absorption and porosity [23]. Nanocomposites were observed to enhance the dielectric response by the addition of a very small amount of an inorganic filler, whose k is almost equal to that of a polymer [27]. For a polymer matrix, the percolation threshold was found to depend mainly on the filler shape and dimension [43]. Studies have reported on that nanorod composites could easily achieve the percolation threshold [20]. The nanofiller approach increases the k value of polymer composites, but it affects the dielectric strength E_b of a composite material. The existence of an inhomogeneous field distribution owing to the large difference in k values of a ceramic nanofiller and the polymer matrix which yielded sharply decreasing E_b of the nanocomposites [7]. BaTiO₃/PP and TiO₂/PP achieved a high energy density of 9.4 J/cm³. Rod-shaped TiO₂ exhibited a high k and a low breakdown strength than spherical ones [16, 38]. It was observed that spherical nanoparticles of BaTiO₃ could be used for enhancing the electrical polarization, while hexagonal boron nitride nanosheets and MMT nanoplatelets were more suitable for increasing the dielectric breakdown strength [31]. M. Chun reported on that, as the size of BaNd₂Ti₄O₁₂ filler decreases from 2.45 μm to 0.69 μm, the dielectric constant and tan δ decrease from 7.2 to 4.8 and 0.0068 to 0.0052, respectively. This variation of the dielectric constant and dielectric losses with the filler size is plotted in Fig. 2 [31].

3.5. Filler loading

An increase in the filler content greatly increases the dielectric constant. But the large content of a ceramic filler makes the composite bulky and increases the loss of flexibility and results in the poor dispersion of a filler [3, 24]. The loading of a filler affects the dielectric loss and strength of a composite as

well. B.K. Sharma *et al.* showed that the loss factor decreased with the addition of ZnO in the polyaniline (PANI) matrix [46]. Y. Deng *et al.* reported on that the dielectric loss increased with the loading of Bi₂S₃, as Bi₂S₃ is a semiconductor [20]. At a low filler loading, the composites exhibited high k and a high dielectric strength. Incorporating metal oxide BaTiO₃ and TiO₂, the high energy composite ($U = 9 \text{ J/cm}^3$) was obtained with no influence on the dielectric strength of PP [47]. Figure 3 shows a variation of the dielectric constant with the filler loading with reference to the theoretical model.

Table 1 shows the effect of filler loading on different polymer composites:

3.6. Composite microstructure

High dielectric constant can be achieved by reinforcing the polymer matrix by a filler with high dielectric constant or by a higher loading of the filler. A reduction in the size of a filler improves the dielectric constant of composites. But it comes with associated problems such as: at a higher loading, the material becomes bulky, and there occurs a deterioration of mechanical properties. Nanoparticles have a tendency to agglomerate. In addition, due to the formation of a local field, the dielectric strength of a composite reduces significantly. Nanocomposites possess high moisture absorption and porosity resulting in high dielectric losses. The large interface volume of a nanocomposite causes an increase in the loss factor and a decrease in the dielectric strength [9]. With the objective of designing a composite at a lower loading with maximum extractable energy, a hybrid composite can be produced with a proper core-shell structure. Results have been reported with high k , low loss and high energy density with hybrid composite approach. M. Roy *et al.* showed that, for microfilled composites, the dielectric strength is less than for a nanofilled composite in polyethylene/SiO₂ [37]. T.S. Sasikala *et al.* showed that nanocomposites exhibit higher loss as compared with microcomposites in Mg₂SiO₄/PTFE [23].

3.7. Surface treatment

Interface compatibility of an inorganic filler and an organic polymer is very weak, since their properties are completely different, which affects the dispersion of a filler in the matrix. The surface treatment of filler particles or the core-shell structured

approach significantly enhances the dielectric constant, reduces the loss factor, and decreases the frequency dependence [32]. The homogeneous dispersion of a nanofiller can be achieved even at a high loading. Simultaneously, the dielectric constant of a composite can be effectively enhanced with a reduction in the dielectric loss and maintaining the dielectric strength at the same time [58, 59]. Zhou showed that the dielectric constant of AlN can be improved with surface treatment [29]. Cheng Yang *et al.* reported on that multi-walled carbon nanotubes (MWCNT) treated with polypropylene show a good dispersion in polystyrene and a reduced leakage current [53]. In PS/MWCNT composite, bare carbon nanotubes (CNT) lead to a huge dielectric loss. MWCNT treated with polypropylene show a good dispersion in polystyrene and a reduced leakage current [53]. Ceramic fillers increase the density and porosity of composites with loading. At a low porosity, the uniform dispersion and good adhesion are observed between a ceramic filler and the polymer matrix. T.S. Sasikala and M.T. Sebastian reported on that silane-treated composites showed a lower porosity [23]. The silane treatment and the coupling agent formed a strong bond between the organic and inorganic phases. K.S. Shah *et al.* showed that the silane-treated montmorillonite clay composite not only showed a higher k value, but also a higher dielectric strength [55]. Three-phase composites restricted a decrease in the dielectric strength due to the addition of nanofillers [6]. Another study reported on that the Al₂O₃ shell layer reduced the interfacial polarization, electrical conduction, and electric field concentration which, in turn, enhances the dielectric strength and decreases the loss factor [59].

3.8. Methods of experimentation

Three general methods are used to disperse a nanofiller in the polymer matrix. A) direct mixing B) in situ polymerization in the presence of nanoparticles, and C) in situ formation of nanoparticles with simultaneous polymerization [3, 28]. In the conventional blending of the two components, the method raises the issues of inhomogeneity and aggregation [60]. In first two methods, filler particles do not disperse uniformly in the matrix polymer, as the polymerization of organic monomers and the formation of guest particles are separately carried out [28]. The in situ formation limits the size of nanoparticles

Table 1. Effects of filler loading on polymer composites

Sr. No.	Polymer	Filler	Feature
1	Silicon	Polyethylene glycol (PEG)	Addition of filler content in matrix increased k value and loss factor [24]
2	Polyaniline (PANI)	Fe_3O_4	k value and loss factor both decreased with loading of Fe_3O_4 [48]
3	Polymethyl vinyl siloxane (VMQ)	Hexagonal boron nitride(HBN)	k value and loss factor improved with increasing HBN content [49]
4	UHMWPE/LLDPE blend	$\text{Al}_2\text{O}_3/\text{Si}_3\text{N}_4$	k value and loss factor decreased with loading [50]
5	Polyvinylidene difluoride (PVDF)	Al	k value increased with loading and no variation was observed in loss factor upto 60 wt% of filler [51]
6	Polystyrene (PS)	TiO_2	With loading of filler, k value increased and loss factor decreased [43]
7	PS	BaTiO_3	Addition of filler content in matrix increased both k value and dielectric loss [32]
8	Cyclic olefin copolymer (COC)	BST	Gradual increase in k value and loss factor with BST loading was observed [52]
9	PS	MWCNT@PPy	Increasing wt% of MWCNT increased k and loss factor but breakdown strength was decreased [53]
10	PVDF	SiC-CNT	Loading of filler increased k value and loss factor. Highest value of $k = 8700$ was obtained at 2.3 vol% of CNT [22]
11	PS	BaTiO_3 -PMMA	k value increased with loading of BaTiO_3 . Highest extractable energy 2 J/cm^3 was obtained at field strength E_b 220 kV/mm for 22 vol% BaTiO_3 [54]
12	Polytetrafluoroethylene (PTFE)	Mg_2SiO_4	k value and loss factor both were increased with loading of filler [23]
13	Polypropylene (PP)	BaTiO_3 , TiO_2	At low volume fraction of inorganic filler, k value of nanocomposite moderately increased with loading [16, 38]
14	High Density Polyethylene (HDPE)	Montmorillonite clay	Increase in clay content increased the k value. Dielectric strength increased upto 5 wt% filler content [55]
15	Cynoresin (CRS)	TiO_2	Nanocomposite showed ultrahigh k which was found to increase with loading. Loss factor remained low <0.3 [30]
16	PVDF	Boron nitride nanosheet (BNNS), Barium Strontium Titanate (BST)	This is trilayered composite. Increase in BNNS content beyond 10 vol% resulted in systematic decrease in dielectric strength. Both k and loss factor increased with loading of BST. Superior power density 0.91 MW/cm^3 more than BOPP [7]
17	Cynoethylated Cellulose (CRS)	Montmorillonite clay	Loading above 5 wt% showed that composite exhibited high k value but decreased with clay loading [56]
18	PVDF-HFP	PFBPA- BaTiO_3 (Pentafluorobenzyl phosphonic acid bounded on BaTiO_3)	Increased filler content decreased the dielectric strength. k value increased with loading and attained maximum at 50–60 vol% of filler. Beyond this k value showed a decrease [12]
19	Benzocyclobutane (BCB)	BNNS	Dielectric strength of composites improved to 403 MV/m on addition of 10 vol% BNNS in matrix [57]

and prevents their agglomeration [3]. In the recent years, the third component in two-phase composite systems is introduced. Y. Li *et al.* obtained a high value of the dielectric constant by adding the third

component in a two- phase PVDF composite, i.e., for PVDF/BT/ β -SiC, $k(\text{high}) = 325$ with 40 vol% BaTiO_3 and 17.5 vol% β -SiC [34]. The dielectric constant of PDMS(3.1) was improved to 5.5 at 1 GHz by

introducing core shell Fe covered by $\text{SiO}_2(\text{Fe-SiO}_2)$, and k was increased to 9.8, when Fe was covered by $\text{TiO}_2(\text{Fe-TiO}_2)$ [61].

3.9. Porosity

Porosity is one of the prime factors affecting the dielectric properties of inorganic and polymer-based composites. In ceramics, the porosity is not desirable, as usual. The porosity is minimized, and the permittivity and dielectric losses of ideal fully dense ceramics are estimated by the porosity. The porosity reduces the breakdown voltage, by resulting in the degradation of the electrical reliability performance of a dielectric material, but does not affect the leakage current. It can be concluded that the use of the fillers with a low dielectric constant or increased porosity of a material lowers the dielectric constant of the composite material [9].

3.10. Dielectric properties of polymer composites

Most of the commercially available electrostatic capacitors possess energy densities in the range of 18 to 29 J/cm³. Biaxially oriented stretched polypropylene (BOPP) is the best dielectric material currently used in electronic circuits due to its high dielectric strength (>700 MV/m) and low dielectric losses (<0.02%). BOPP possesses $U = 2 \text{ J/cm}^3$ with k value 2–3 [36]. Polypropylene is a tough, crystalline, rigid, the lightest, and cheapest thermoplastic polymer. It has excellent piezoelectricity [47]. It possesses a high dielectric strength of $\sim 200 \text{ MV/m}$ [3]. N. Guo *et al.* synthesized polypropylene (PP) composites with BaTiO_3 , TiO_2 , ZrO_2 and showed that BaTiO_3 composite possesses the highest energy ($U = 9.4 \text{ J/cm}^3$) at 13.6 vol% BaTiO_3 [16]. But the use of PP is limited due to a low dielectric constant [36]. High dipole density polymers provide a potential to increase the energy density. Poly(vinylidene fluoride) (PVDF) and its copolymer trifluoroethylene (TrFE) are polar-polymers, the best known ferroelectric polymers. These polymers have been widely used in actuators and electro-mechanical sensors [8]. PVDF is a tough, stable, semicrystalline thermoplastic ferroelectric polymer. PVDF has low density in comparison with other fluoro-polymers. PVDF exhibits the high chemical resistance and compatibility among thermoplastic materials. PVDF has sturdy tensile strength and contains two fluorine groups which show an in-

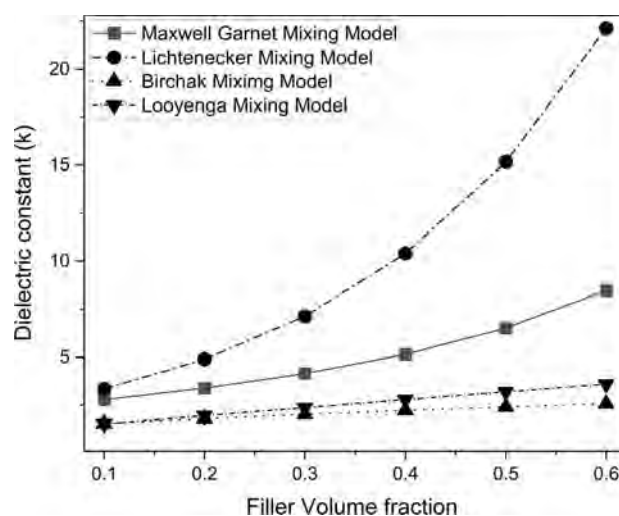


Fig. 3. Variation of the dielectric constant with filler volume fraction

tense polarity [47]. Zhou showed that the Al filler decreases the degree of crystallinity of composites. High k value ($k = 230$) is obtained at 80 vol% of Al [29]. The high breakdown strength of PVDF has also been investigated by researchers introducing 3D ZnO superstructure [52]. The large remnant polarization in normal PVDF and PVDF-TrFE causes a low energy density. A study reported on the modified PVDF-TrFE to ferroelectric relaxor in which remnant polarization remained zero and acquired large energy density $> 9 \text{ J/cm}^3$ and high dielectric strength 400 MV/m. PVDF-TrFE(91/9 mol%) shows $U > 17 \text{ J/cm}^3$. PVDF and its copolymers promise a high energy density, but suffer from critical property defects such as the high dielectric and hysteresis losses, poor thermal stability, dielectric losses depending on the frequency and temperature, *etc.*, which limits their use in industrial applications [17]. Composites of polystyrene show the almost frequency-independent dielectric behavior for BaTiO_3 , TiO_2 , and MWCNT-PP fillers [43, 53, 54]. Polar polymers possess a good affinity for inorganic oxide fillers. Researchers use cynoresin for the development of dielectric polymer composites due to its polar backbone. N. Madusanka *et al.* showed that TiO_2/CRS possesses a high dielectric constant of 207 at 30 vol% of TiO_2 [56]. Montmorillonite clay which is non-toxic and cheap increases the dielectric constant of CRS up to 10 vol% of MMT ($k = 49$) [56]. Dielectric Properties of some polymer composites are listed in Table 2.

Table 2. Dielectric properties of polymer composites

Polymer	Filler	k/U value	$\tan \delta/E_b$	Ref.
PVDF	SiC-CNT (2.3 vol%)	$k = 8700$	$\tan \delta = 286$	(22)
	Al (80 wt%)	$k = 230$	$\tan \delta = 0.25$	(29)
	BaTiO ₃	$k = 400\text{--}450$	–	(41)
	BaTiO ₃ @Al ₂ O ₃	$k > 10$	$\tan \delta = 0.02$	(59)
	x GnP (exfoliated Graphite nanoplates)	$k > 200$	$\tan \delta = 229$	(21)
	ZnO (commercial)	$k = 19.4$	$E_b = 45$ kV/mm	(50)
	ZnO (flower like)	$k = 221.1$	$E_b = 42$ kV/mm	(50)
	ZnO (walnut like)	$k = 104.9$	$E_b = 40$ kV/mm	(50)
	Ba(Zr _{0.3} Ti _{0.7})O ₃ (BZT)	$U = 2.8$ J/cm ³	$E_b = 400$ kV/mm	(62)
	PVP modified BZT	$U = 6.3$ J/cm ³	$E_b = 380$ kV/mm	(62)
	β -SiC	$k = 260$	$\tan \delta = 1.46$	(34)
	BaTiO ₃ / β -SiC	$k = 325$	$\tan \delta = 0.36$	(34)
	BaTiO ₃ @SiO ₂	$k = 31.12\text{--}41.65$	$\tan \delta = 0.014\text{--}0.028$	(63)
	TiO ₂ @BaTiO ₃	$U = 12.2$ J/cm ³	$E_b = 340$ kV/mm	(58)
	BaTiO ₃ @TiO ₂ nfs	$k > 41, U = 20$ J/cm ³	$E_b = 650$ kV/mm	(44)
	BaTiO ₃	$k \sim 9.20\text{--}22.02$	$E_b \sim 174\text{--}240$ kV/mm	(15)
	BNNS	$U \sim 12.5$ J/cm ³	$E_b = 672$ MV/m	(7)
	BST	$U \sim 10$ J/cm ³	–	(7)
	Trilayered (PVDF-BST/PVDF-BNNS/PVDF-BST)	$U \sim 20$ J/cm ³	$E_b = 588$ MV/m	(7)
			$\tan \delta < 0.05$	(7)
P(VDF-TrFE)	CaCu ₃ Ti ₄ O ₁₂	$k = 245\text{--}362$	$\tan \delta = 0.32\text{--}0.42$	(1)
P(VDF-HFP)	(PFBPA-BT)	$U = 3.2$ J/cm ³	$E_b > 164$ kV/mm	(12)
P(VDF-CTFE)	BNNSs/BT	$k \sim 8.0\text{--}15.3$	$E_b \sim 649$ kV/mm	(13)
	BST	$U = 6.5$ J/cm ³	$E_b > 200$ kV/mm	(14)
PP	Al ₂ O ₃ @Al	$U = 13.4$ J/cm ³	$E_b = 75.8$ kV/mm	(58)
	PVDF (blend)	$k = 3$	$\tan \delta < 0.2$	(47)
	SMA/PVDF (blend)	$k = 13$	$\tan \delta < 0.03$	(47)
	Al ₂ O ₃	$k < 5$	$\tan \delta < 0.040$	(35)
	BaTiO ₃ (BT)	$k = 20$	$\tan \delta < 0.050$	(35)
	Al ₂ O ₃ /BT	$k = 18$	$\tan \delta < 0.030$	(35)
	BaTiO ₃ /TiO ₂	$U = 9.4$ J/cm ³	$E_b \sim 4$ MV/cm	(38)
	TiO ₂	$k \sim 4.9$	$E_b > 2.8$ MV/cm	(16)
			$U \sim 0.2\text{--}2.8$ J/cm ³	
	BaTiO ₃	$k \sim 6.1$	$E_b > 5.9$ MV/cm	(16)
			$U = 9.4$ J/cm ³	
	ZrO ₂	$k \sim 6.9$	$E_b > 2.0$ MV/cm	(16)
			$U = 1.02$ J/cm ³	
PS	BaTiO ₃	$k \sim 2.76\text{--}24.51$	$\tan \delta \sim 0.013$	(32)
	TiO ₂ (11.9 wt%)	$k \sim 10$	$\tan \delta < 0.05$	(43)
	TiO ₂ (36.9 wt%)	$k \sim 80$	$\tan \delta < 0.15$	(43)
	TiO ₂ (27 wt%)	$k \sim 6.4$	$\tan \delta \sim 0.04$	(60)
	PMMA-BaTiO ₃	$U = 2$ J/cm ³	$E_b = 220$ kV/mm	(54)
	MWCNT@PP(3%)	$k = 4.3$	$\tan \delta \sim 0.005$	(53)
	MWCNT@PP(3%)	$k = 44$	$\tan \delta \sim 0.02$	(53)
LLDPE	AlN (50 wt%)	$k = 5$	$\tan \delta < 0.085$	(29)
	AlN (70 wt%)	$k = 6.9$	$\tan \delta < 0.085$	(29)
UHMWPE/LLDPE	Si ₃ N ₄	$k = 3.28$	$\tan \delta = 4.68$	(50)
	Al ₂ O ₃ /Si ₃ N ₄	$k = 3.19$	$\tan \delta = 4.49$	(50)
LDPE	Bi ₂ S ₃	$k = 60$	–	(20)

3.11. Application of dielectric polymer composites

High dielectric constant materials are used to design high energy storage capacitors, pulsed power capacitors, gate dielectrics, epitaxial dielectrics, and dielectric elastomers [3, 45, 64]. Low dielectric constant polymer composites are used in the electrical insulating and packaging materials [3, 11]. Microwave dielectric materials have a wide range of applications such as the satellite communication and straddling terrestrial ones, including software radio, Global Positioning System (GPS), Direct Broadcast Satellite (DBS) TV, Internet of Things (IoT), environmental monitoring via satellites, dielectric resonators, substrates for microwave integrated circuits, etc. Tunable dielectric materials are used in antenna beam steering [5, 9].

4. Conclusion

- Achieving a high dielectric strength with improved dielectric constant is a major challenge.

- Inorganic ceramic fillers improve the dielectric constant of polymer composites; however, they reduce the dielectric strength and mechanical properties.

- The uniform dispersion of a filler and the low percolation threshold play a vital role in improving the dielectric properties.

- Core-shell structured and multi-layered composites ensure the improvement of the dielectric constant and a high dielectric strength at a low-level loading.

- To achieve a high energy density, materials should possess high k and high dielectric strength values.

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Б. Гхуле, М. Лаад

ПОЛІМЕРНІ КОМПОЗИТИ З ПОЛІПШЕНИМИ ДІЕЛЕКТРИЧНИМИ ВЛАСТИВОСТЯМИ: ОГЛЯД

Матеріали з високим значенням діелектричної константи (k) застосовуються в конденсаторах, як ізолюючий шар у затворах, в діелектричних еластомерах, акумуляторах енергії, тоді як матеріали з малим k потрібні для електронного монтажу і в багатьох інших випадках. Традиційно матеріали з високим значенням k характеризуються значними діелектричними втратами, залежністю властивостей від частоти і великим навантаженням на наповнювач. Матеріали з малим k мають низьку теплопровідність. Все це створює нові завдання у розробці діелектричних матеріалів для обох видів застосування. Використання наповнювачів з високим значенням діелектричної константи дає збільшення сумарної k . Ми вивчаємо вплив різних параметрів і факторів на діелектричні властивості матеріалів, а саме: тип полімеру, матеріал наповнювача, розмір, форма, рівень навантаження, модифікація поверхні матеріалу наповнювача і спосіб приготування полімерного композита. Роботу спрямовано на поліпшення діелектричних характеристик полімерних нанокомпозитів, які застосовуються у пристроях для накопичення енергії. Результати показують, що підхід зі структурованими ядром і оболонкою для матеріалів з великим k , введеними в полімерну матрицю, дозволяє покращити діелектричні властивості полімерних композитів.

Ключові слова: полімерні композити, діелектрична константа, електрична міцність діелектрика, керамічний наповнювач, накопичення енергії.