The Future of Nanodielectrics in the Electrical Power Industry

Yang Cao, Patricia C. Irwin and Karim Younsi
GE Global Research Center
Niskayuna, NY, 12309 USA

ABSTRACT

While specialty applications of nanotechnology in the photonics and electronics areas have seen a tremendous growth in the past several years, the use of nanodielectrics in the electrical industry (high power density and high voltage) has not shown the same level of activity. In addition to a review of nanodielectrics, we discuss in this paper, our perspective on the current status, development needs and future potential to build or engineer nano-structured materials for dielectric applications in the electrical power industry. Short and long-term future research and development needs are considered from the point of view of industrial applications.

Index Terms — **Nanodielectrics, nanocomposites, nano structure, dielectrics, electrical insulation.**

1 INTRODUCTION

NANODIELECTRICS is the study of dielectric phenomena and materials on the nanometric scale and the fabrication of structures, devices and systems that have novel dielectric properties and function because of their nanometric structure [1, 2]. The novel and differentiating properties of these structures are developed at a critical scale of matter, typically under 100 nanometers. The scope of nanodielectrics however, can be broad and extensive [2, 3]. Therefore, in this paper, we will use "simple" two-phase heterogeneous nanodielectric systems as examples to discuss the long and short-term development needs and applications of nanodielectrics.

Currently, the synthesis and fabrication of nanometer-sized particles have reached a mature state and they can be made with controlled composition and size [1]. Because of their length scale and high specific surface area, nanoparticles exhibit novel properties as compared with bulk materials. A lot of research has been carried out recently incorporating various nanoparticles into existing dielectric systems in a cost effective manner, resulting in nanocomposites with improved benefits over conventional filler systems [4–7]. The improvements can be a combination of electrical, mechanical and thermal enhancements [5, 6], and can be further tailored through organic synthesis, material design optimization. These types of "top-down" research styles will continue to be fruitful with many important engineering applications in the near future.

Meanwhile, a great deal of progress has been achieved in fabricating and assembling nanostructures to high precision and reproducibility, producing interesting optical, photonic, electronic, and chemical properties [8–13]. While some of the nano structures are formed using dielectric methods [14, 15], the dielectric properties of these novel structures have not been fully explored. These "bottom-up" structures not only provide unique opportunity for the study of dielectric phenomena at the nanometric level, but also have great potential for groundbreaking application of nanodielectrics in the long term. As an example, a giant dielectric constant of $\sim 10^{10}$ was reported for a nano silver assembly in a polymer matrix as a result of quantum confinement of the electronic wave function [16]. It is not difficult to speculate that its potential applications in energy storage and ultra sensitive transducers are tremendous.

2 CURRENT INDUSTRIAL APPLICATIONS

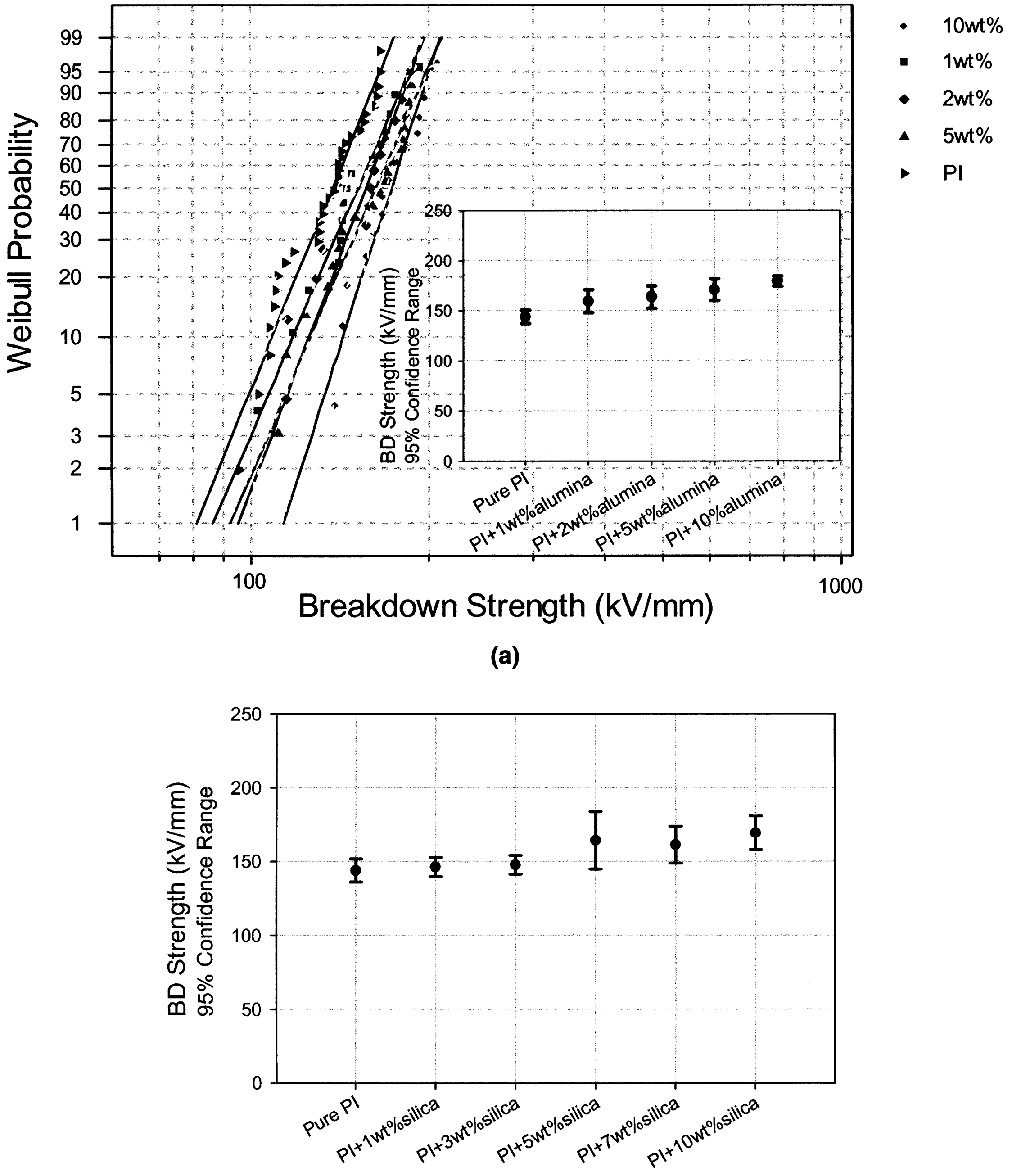
Two-phase heterogeneous nanodielectrics, generally termed as dielectric nanocomposites, have wide applications in the electrical and electronic industries. Various types of nanocomposites, based on insulating, semiconducting or metallic nanoparticles, have been developed to meet the requirements of specific applications. For instance, nanocomposites with anisotropic electrical properties are desirable for improving partial discharge resistance of the ground wall insulation of rotating machinery.

Manuscript received on 21 April 2004, in final form 24 June 2004.

2.1 ELECTRICAL APPLICATIONS

Insulation integrity is of great importance for all-electrical power applications, including energy conversion, power delivery, energy storage, and power consumption [17–20]. Nanodielectrics can enhance the reliability of current systems, and more importantly, can improve their efficiency by enabling innovative design and the utilization of renewable energy resources.

With the ever-growing demand for compact and highly efficient electric machines, the electrical insulation systems are subjected to multiple stresses [21] with greater intensity, and higher repetition rate [18]. While conven-



(a)

(b)

Figure 1. a, 2-parameter Weibull statistic plot of the breakdown strength of polyimide (PI) nanoparticle composites at different loadings of alumina nanoparticles. The tests were performed with a 500 v/s ramp rate. The samples were 25 μm thick. The inset shows the Weibull characteristic breakdown strength vs composition with loadings up to 10wt%. The bars in the inset correspond to the 95% Weibull confidence limits. From the plots, slightly increase of the breakdown strength of PI with loading is observed; b, Breakdown strength plot for PI silica nanoparticle composites tested under the same conditions.

tionally filled polymeric insulation offers improved electrical discharge resistance, thermal expansion coefficient adjustment, mechanical robustness and heat dissipation, it is often achieved at the cost of reduced dielectric performance [17, 19, 20]. The operational field strength is the fundamental design limitation for high voltage systems. It is possible to push back this design barrier by utilizing nanodielectrics. Several typical nanocomposite systems for application in this area, which illustrate the current situation, will now be discussed.

2.1.1 NANOCOMPOSITES WITH INSULATING NANOPARTICLES

An important type of nanocomposite is one with insulating nanoparticles such as silica and alumina. As shown in Figure 1, the electrical breakdown strength of a polyimide (PI) is not affected by these types of nanofillers for loadings up to 10wt%. The study of the conduction mechanism of these nanocomposites [5] indicates a field enhanced, thermally assisted ionic-hopping conduction current

$$j = 2nd\,\nu \exp\left[-\frac{U}{kT}\right]\sinh\left[\frac{eEd}{2kT}\right] \qquad (1)$$

as described earlier [22–24], where U is the activation energy, d is the hopping distance, and ν is the attempt-to-escape frequency, n is the carrier density, k is the Boltzmann constant, T is the temperature and e is the electronic charge. Figure 2 gives a 3D curve fitting of the steady-state current density of 10wt% silica filled PI nanocomposites to equation (1). Normally the addition of conventional micro sized fillers would result in increase of

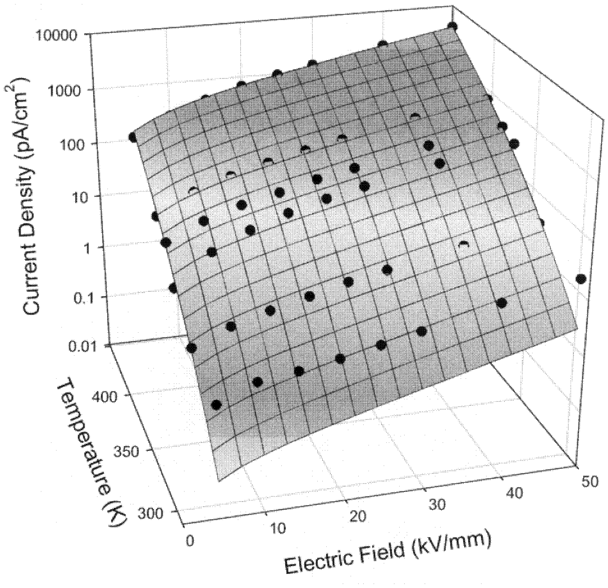


Figure 2. Field and temperature dependence of the steady-state current density for a PI nanocomposite with 10wt% nano silica. The 3D curve fitting of the data to equation (1) gives an activation energy of 0.68 eV and a hopping distance of 4.3 nm [5].

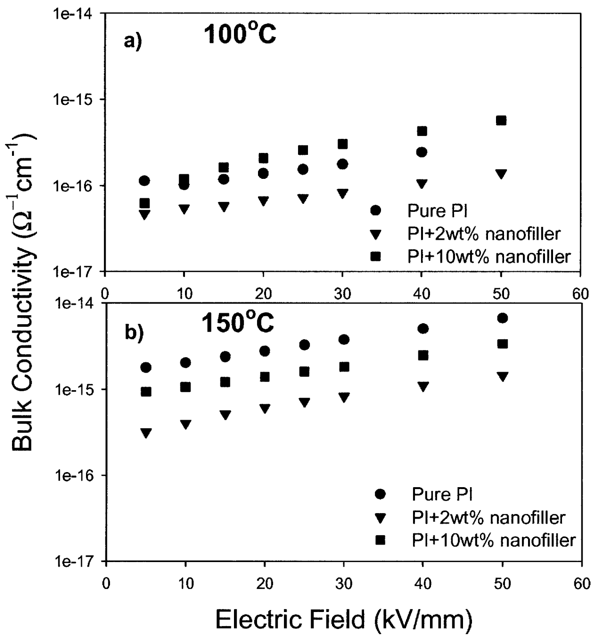


Figure 3. Comparison of the conductivity of pure and nano-filled polyimides. a, conductivity at 100°C; b, conductivity at 150°C. Circles denote the bulk conductivity of unfilled polyimides. 2wt% silica nanofilled nanocomposites show a significant reduction in conductivity at both 100°C and 150°C (diamond symbols) whereas 10wt% nanofilled composites are less dramatic in conductivity changes (square symbols). More work is needed to understand the complex nature of temperature and filler loading effects on the bulk conductivity of a nanocomposites.

bulk conductivity due to the increased defects and poor filler/polymer interactions. Interestingly it is found that 2wt% silica nanofiller shows a significant reduction in conductivity at both 100°C and 150°C whereas 10 wt% nanofilled composites are less dramatic in conductivity changes, indicating a complex nature of temperature and fill loading effects on the bulk conductivity of a nanocomposites (Figure 3).

In addition to the enhancement of partial discharge or corona resistance (Figure 4), the addition of nanoparticles shows a marked improvement in ductility and yield strength, shown in Figure 5, as well as certain improvement of the scratch resistance and thermal conductivity [6]. The improvement of the mechanical properties is important, because an insulating material might be subjected to constant vibration or abrasion by the double power frequency magnetic force and/or high shear stress under rapid thermal loading [25]. As a result, existing high temperature thermosetting insulation, which is inherently brittle, quite often undergoes void/crack formation or delamination under these mechanical stresses with subsequent electrical discharge and catastrophic failure.

Overall, these nanocomposites show combined electrical, mechanical and thermal improvements over conventional filler systems.

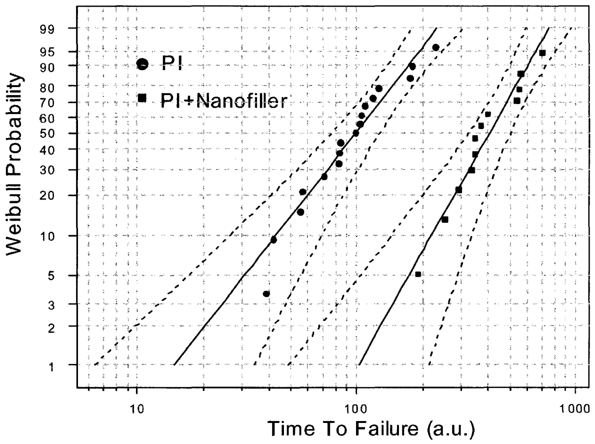


Figure 4. The enhancement of partial discharge resistance of PI with a nanofiller at 10 wt% loading. The dotted lines represent the 95% confidence limits.

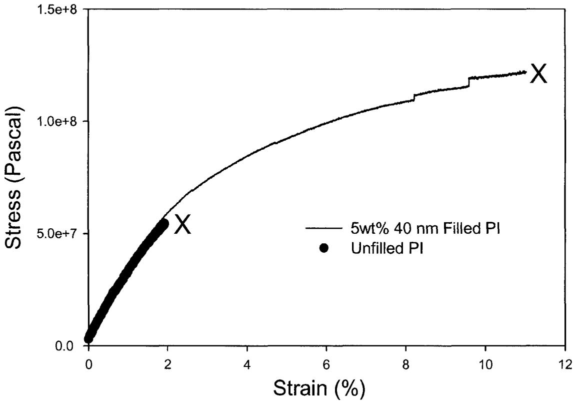


Figure 5. Example of tensile experiments showing an increase in ductility of a polymeric system through the addition of nanoparticles. "X" denotes a mechanical failure during the tensile stress test. An unfilled polyimide fails at 2% strain and 6×10^7 Pa whereas a 5wt% alumina filled PI does not fail until 11% strain and 1.2×10^8 Pa. The Young's modulus of the two materials is initially similar but the nanofilled sample showed significant increase in elongation prior to failure.

2.1.2 NANOCOMPOSITES WITH NONLINEAR NANOPARTICLES

Even though the short term breakdown strength is reduced for nanocomposites with nanofillers shown in Figure 6 (TiO_2, ZnO, $BaTiO_3$, SiC), they form an important class of dielectric materials with special dielectric properties. For instance, SiC and $BaTiO_3$ composites have been employed as field grading materials and ZnO is an excellent surge arrestor additive because their electrical properties (conductivity and/or permittivity) are strongly field dependent. Recent studies indicate that nanocomposites based on these materials have unique dielectric properties [26, 27]. LDPE/ZnO nanocomposites exhibit a lower percolation threshold than classical percolation theory, which describes a power law dependence of conductivity on filler concentration p above a percolation threshold of p_c [26]

$$\sigma \propto (p - p_c)^n \tag{2}$$

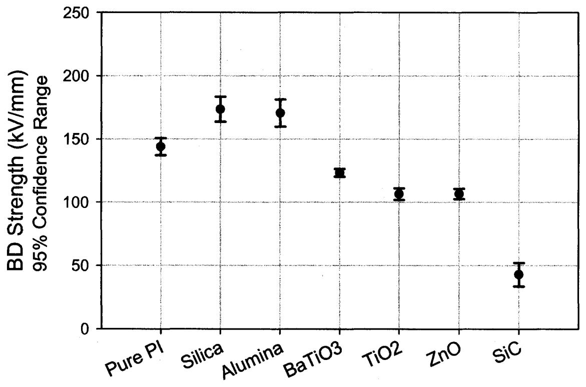


Figure 6. ac breakdown strengths for PI with various nanofillers at 5wt% loading. The tests were performed with 500 v/s ramp rate on samples of 25 μm thick. The remarkably low breakdown strength of SiC nanocomposites may be caused by the aggregation of SiC nanoparticles.

Further investigation shows that for polyethylene filled with micro and nano ZnO fillers, the decrease in resistivity starts at the same critical interparticle distance l_c of about 40nm (Figure 2 of [26]). The critical interparticle distance can be written as

$$l_c = r\left[\left(\frac{4\pi}{3}p_c\right)^{1/2} - 2\right] \tag{3}$$

by assuming a spherical particle shape with a radius of r, and a uniform size distribution and dispersion. Below the critical interparticle distance l_c, LDPE/ZnO nanocomposites exhibit a smaller decrease in resistivity with filler concentration when compared with conventional composites [26].

The addition of conventional TiO_2 filler to polyethylene has been investigated for DC power transmission applications [28], in which the space charge accumulation due to the large thermal gradient across the cable may cause catastrophic failure of the cable insulation system during polarity reversal. It was also shown recently that TiO_2 nanoparticles could mitigate such space charge accumulation in an epoxy system, as compared with conventional TiO_2 fillers [27].

2.1.3 ANISOTROPIC NANOCOMPOSITES

Anisotropic materials and structures inherently have unique properties and applications. Polymer/layered silicate nanocomposites represent good examples of such materials with attractive properties, including high mechanical moduli, heat resistance, barrier behavior, ion-exchange capability. A comprehensive review of the preparation, characterization, properties and processing of polymer/layered silicate nanocomposites can be found in [29]. Technologies have been developed so that layered silicates can be incorporated into most types of polymers and nanocomposites can be formed through melt-mixing

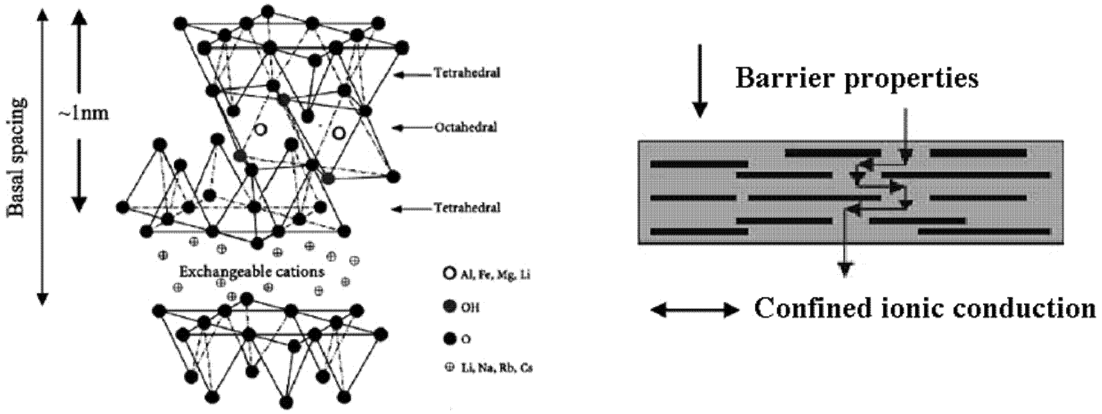


Figure 7. Chemical structure of phyllosilicates, a typical nanoclay whose crystal structure consists of layers made up of two tetrahedrally coordinated silicon atoms fused to an edge-shared octahedral sheet of either aluminum or magnesium hydroxide [29]. The layer thickness is about 1 nm, and the lateral dimensions of these layers may vary from 30 nm to more than several microns. There are three different types of thermodynamically achievable polymer/layered silicate nanocomposites, namely, intercalated, intercalated-and-flocculated and exfoliated [29]. With proper orientation, such layered silicate nanocomposites have remarkable anisotropic properties as shown schematically, i.e., vertical barrier (corona resistance) properties and lateral energy dissipation through confined ionic conduction. Such anisotropic electrical and thermal properties are of great use for rotating machine insulation.

[30]. As shown in Figure 7, the barrier behavior of their layered structure and the adjustable anisotropic ionic conductivity between the layers [31] for such polymer/layered silicate nanocomposites make them particularly interesting for electrical applications.

Mica based insulation systems are used extensively in HV rotating machines for the exceptional partial discharge resistance. The discharge resistance lies perpendicular to the alumino-silicate plane axis due to the strong in-plane bonding. However, the graphitic, layered mica structure can delaminate under thermal-mechanical stresses, forming voids to support partial discharge. As a result, the design field for mica/epoxy or mica/polyester systems is limited to ~ 3 kV/mm [17]. Fully exfoliated and orientated (for example through shear orientation [32, 33]) silicate nanocomposites can potentially be engineered with good discharge resistance and with improved mechanical properties. Moreover, supramolecules with similar structure have been fabricated in the laboratory, targeting anisotropic ionic conduction, for applications such as fuel cells [34].

Carbon nanotubes potentially can find their applications in electric power engineering also. Because of their high aspect ratio, randomly distributed carbon nanotubes can induce significant property change at only 1−5% loading [35]. Compared with conventional carbon black filler, properly distributed and oriented nanotubes within the matrix will lead to many applications, including field grading.

2.2 ELECTRONIC APPLICATIONS

The electronics industry is driving a significant portion of the nanocomposites research and development. With the continuing miniaturization of microelectronic structures, low k dielectrics (below 3 as compared with 4.5 for silicon dioxide) are needed to reduce the signal transition time, power consumption as well as crosstalk through capacitive coupling [36−38]. Nanoporous dielectrics composed of an insulating matrix embedded with pores of nanometer size are candidates. For example, silica aerogels/xerogels having closed pores with porosity as high as 90% while retaining good dielectric properties have been developed [39]. The relative permittivity can be controlled through porosity adjustment and can be as low as 1.3. The challenges lie in the compatibility with current systems and in the mechanical robustness to chemical-mechanical polishing (CMP). Research shows that polymer based nanoporous low-k materials used in a spin-on process compete with modified chemical vapor deposition (CVD) films [40, 41]. In addition to interconnections in the high-density integration, advanced packaging requires also low permittivity dielectrics for RF applications [42]. Meanwhile, polymer-ceramic nanocomposites with high dielectric constant are needed for "System-On-Package" and embedded capacitors applications [43−45].

2.3 OTHER APPLICATIONS

There are many other applications of nanodielectrics such as ionic conductors, nanoceramics for electronics applications [46] and effective EMI/EMC (Electromagnetic Interference/Compatibility) shielding. High frequency EMI/EMC designs require better solutions than conventional metallic shielding in electronic enclosures in order to attenuate unwanted electromagnetic radiation without reflection. For wave propagation in inhomogeneous media, RF "resonant cavity" reflections due to the mismatch of the intrinsic impedances at the enclosure interface are problematic. Dissipative dielectric absorbers with con-

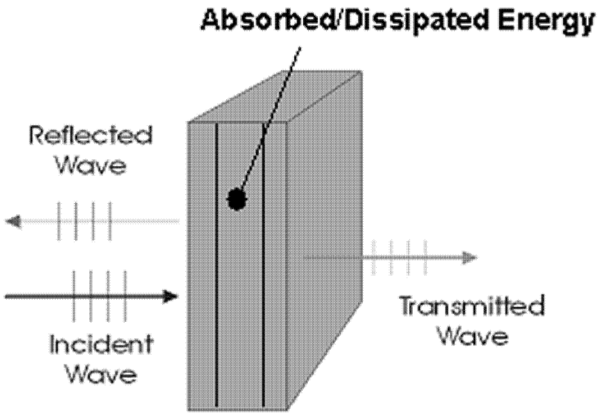


Figure 8. Stealth-like dielectric absorbing materials with controlled permittivity (ϵ), permeability (μ) and conductivity (σ) can offer more flexible EMI/EMC designs than traditional enclosure confinement by attenuating incident electromagnetic radiation. A conventional metallic enclosure can be viewed as a resonant cavity because of reflections by mismatching of the intrinsic impedances ($\sqrt{(\mu/\epsilon)}$). Soft ferrite nanoparticles are useful not only for permeability adjustment but also for the enhancement of electromagnetic absorption at high frequency.

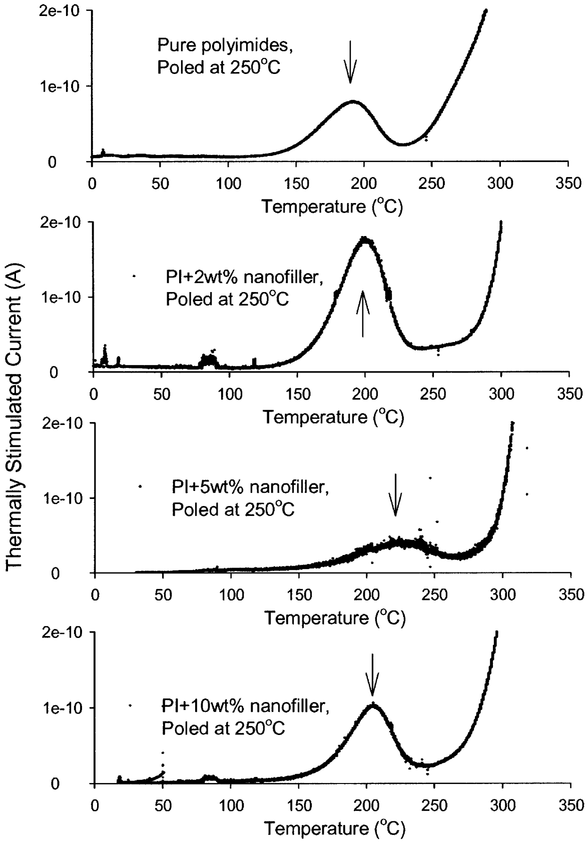


Figure 9. Thermally stimulated current (TSC) data for polyimide nanocomposites with various filler loading. Poling was performed with a field of 20 kV/mm at 250°C for 10 minutes. A heating rate of 2°C/min was used during the TSC acquisition. Since dynamic thermo-mechanical analysis (DTMA) shows no major dipolar relaxation in this temperature range the peaks are likely to correspond to the release of trapped carriers by the thermally stimulated process. The shift of the TSC peak temperature indicates deep trapping of carriers by the incorporated nanoparticles.

trolled permittivity, permeability and conductivity can potentially offer near stealth-like functionality to eliminate undesirable RF reflections, as shown schematically in Figure 8. Such development will have wide applications in the electronics, telecommunications and automotive industries.

3 DEVELOPMENT NEEDS AND FUTURE TRENDS

It would be desirable if nanocomposites could be fabricated with tailored dielectric properties, i.e., controllable permittivity and conductivity as a function of temperature, electric field and frequency. As particles shrink to nanometric sizes the interface becomes increasingly important and ultimately totally dominant. The understanding of the role interface in nanodielectric is far from being satisfactory. Success is limited to a qualitative level for even a simple nanostructure [1, 47] and, quite often, classical approximations such as the effective medium method are used in practice [45]. Therefore, a good understanding of the interaction between nanoparticle and matrix, interplay between particles, and associated transport phenomena is of great importance.

Incorporating particles of nanometric scale has rendered interesting physical, chemical modifications of the matrix dielectric system. Again, if we take silica-filled PI nanocomposites (nanocomposites with insulating nanoparticles) as an example (Figure 9), main thermally stimulated current measurements show clearly the shift of the peak current temperature from 185°C for pure polyimides to about 200°C for polyimides with 2 wt% nanofiller and almost 220°C for polyimides with 5 wt% nanofiller. When the filler loading increases to 10 wt%, the peak temperature reduces to about 210°C. Because a higher peak current temperature usually corresponds to a higher activation energy for trapped charges, deeper trapping is introduced by incorporating nano silica fillers.

In addition to such physical modifications, nanofillers can induce chemical modifications. A nano TiO_2 filler in polyethylene induces a dielectric response by a slight oxidation of the polymer such that a previously not visible dielectric response is seen clearly on the relaxation maps of polyethylene [48]. Dielectric models need to be developed to better describe such physical/chemical modifications by nanofillers as well as the dipolar activities at the interface and the associated transport phenomena.

While these approaches through incorporation of nanoparticles will make progressive improvements in important engineering fields in a cost effective way, they have limitations such as wide particle size distribution, particle agglomeration, etc. Nanocomposites based on semiconductive nanofillers like TiO_2 have been studied extensively not only because of a chemically active surface, but also because of their electronic and optical confinement capability [1]. Semiconductive particles (quantum dots) have been assembled into arrays using pattern-formation block co-polymers as templates [49] and more importantly, their photonic/electronic properties can be altered with the particle size, showing clearly quantization effects in which the energy levels available for electrons and holes become discretized and confined in a nanometer size range. This unique property has led to many applications in electronics, photonics and potentially dielectrics also. In 1965, Gor'kov and Eliashberg, predicted an enormously enhanced polarizability of a sufficiently minute metallic particle having discrete energy levels [50]. However, such prediction had to be corrected for the depolarization field within the particle and the results of high dielectric constant were interpreted using an "interrupted-strand model" - a real crystalline compound consisting of a system of collinear metallic strands, each of which is interrupted by a series of perfectly insulating lattice defects [51]. According to such a strand model, the dielectric constant can be written as

$$\epsilon \approx \frac{1}{2}(q_s l_0)^2 \tag{4}$$

where q_s is the Fermi-Thomas screening wave vector of the conduction electrons and l_0 is the strand length.

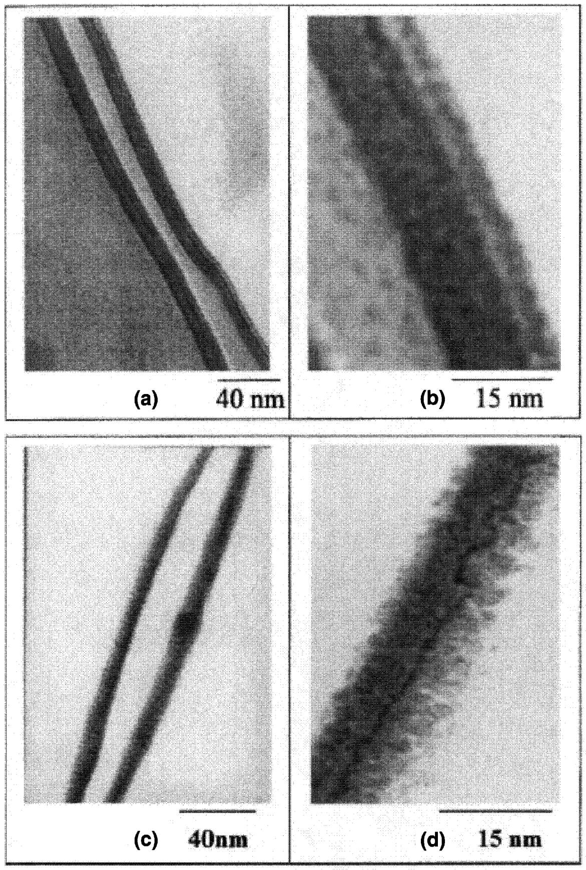


Figure 10. Formation of an assembly of ultrafine metal particles connected to each other to form an interrupted metal strand by the application of electric field to an assembly of isolated minute metal particles. The strong electrostatic force due to the high electric field (10^6 V/cm) in the nanometric air gap between the particles causes them to deform in the direction of the electric field and finally to attain a percolation configuration with the formation of a metal filament. The electronic wave functions of these assembly are localized due to lattice defects at the interfaces between the particles [16]. a, Transmission electron micrograph (TEM) of nanowires; b, diameter of the wire, 15 nm in the magnified view of the wire of (a). From the figure it is evident that the wire is formed by the metal nanoparticles; c, TEM photo of the wire after the application of voltage pulse; d, filamentary growth of ultra fine particles.

Many attempts failed because of the poor control of the structure at the nanoscale [52–56]. It is reported only recently that, using a polymer nano template (with 15 nm pores), researchers [16] were able to fabricate and assemble a silver nanowire as shown in Figure 10. By applying a bias, the wire turns into an assembly of ultrafine metal particles with localized electronic wave function. A dielectric constant of 10^{10} was reported for such a dielectric-

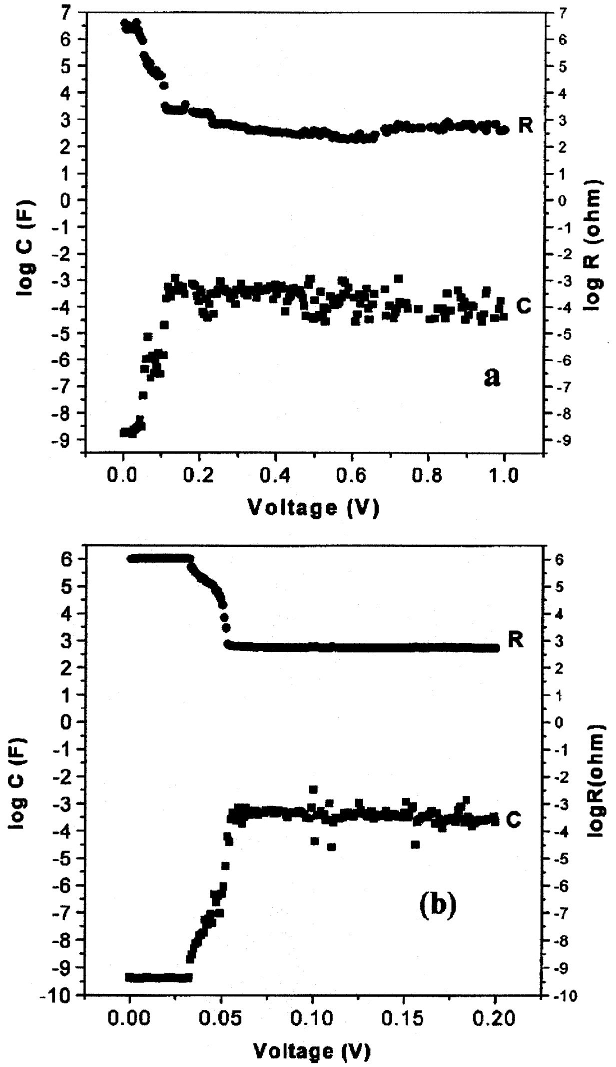


Figure 11. Capacitance and resistance under bias. a, 20 Hz; b, 1 KHz, indicating a giant ϵ of 10^{10} [16].

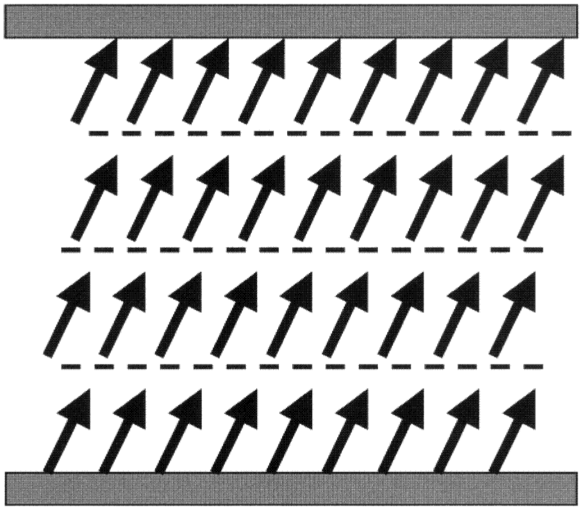


Figure 12. Electrostatic self-assembled monolayer (ESAM) technique for fabricating non-centro symmetric structures [58].

conducting particle assembly [16]. Extremely high dielectric constant materials would be useful as ultracapacitors for energy storage. In addition, because of the surface sensitivity of such a nano assembly and because of its ultra fast response, since it relies on electronic rather than dipolar polarization, it can be developed into an ultra sensitive airborne sensor.

Dielectrics research should also benefit from dramatic progress in nanotechnology where supramolecular structures have been assembled for novel properties [57]. For example, an electrostatic self-assembled monolayer (ESAM) technique for fabricating non-centrosymmetric structures has been reported [58]. Such a technique results in tens of microns thick films with large, stable second order nonlinear optical responses without electric field poling. As shown in Figure 12, a multi-layer film is formed by alternately immersing the substrate in aqueous solutions of a polyanion and a polycation, with one or both the polyanion and polycation containing polarizable chromophores. Similar self-assembly methods have been developed to generate superlattices with oriented giant dipoles for electro-optic applications [59,60]. Nanostructured materials with giant electrostriction as well as organic composite actuator material with high k have also been reported [61,62].

While current research is focused on filled systems, in the long run, nanostructured dielectric materials and devices will have many intentionally designed novel dielectric properties and have wide applications. Figure 13 shows

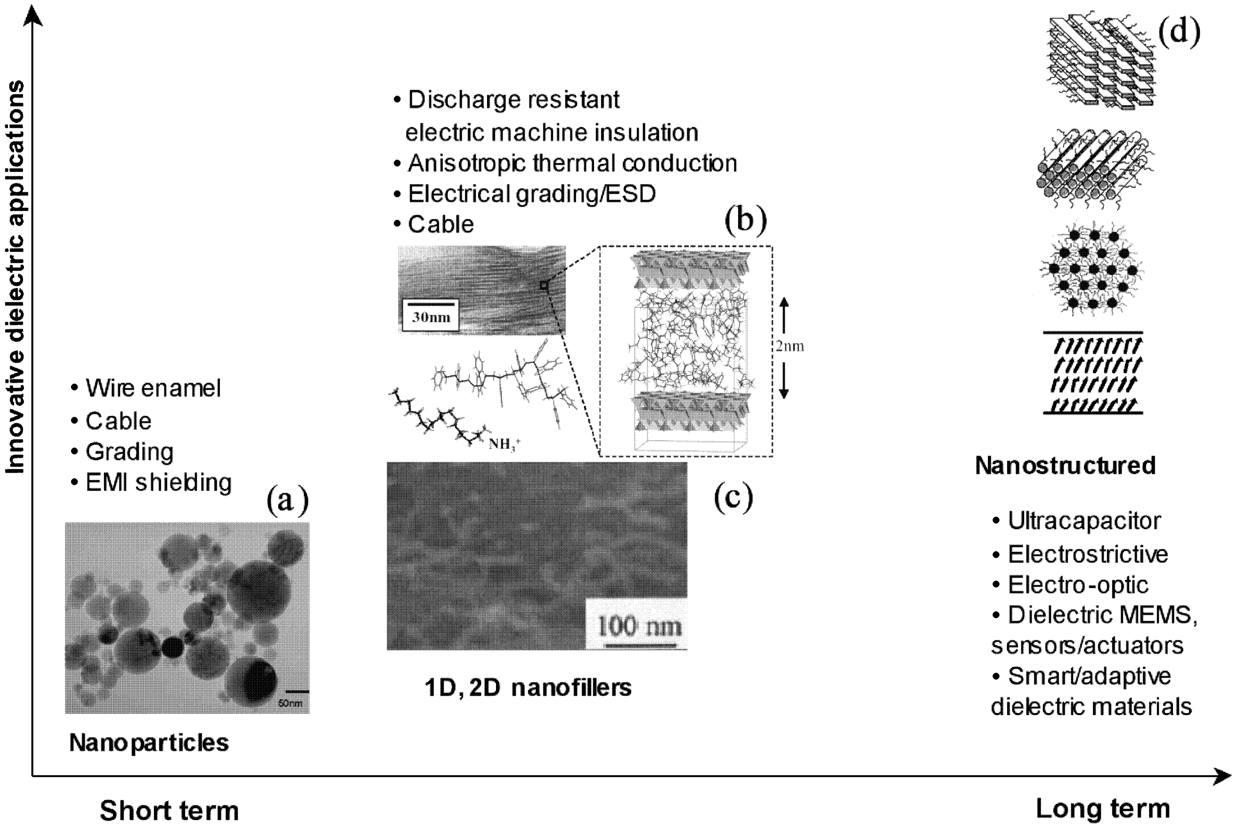


Figure 13. Time scale in the development of nanodielectrics with novel properties/applications for illustration only: a, TEM photo of alumina nano particles dispersion; b, a bright-field TEM detail of the PS/octadecyl-ammonium silicate intercalate and the simulation of the structure: a styrene 12mer and the octadecyl-ammonium surfactant molecule [63]; c, SEM photo of carbon nanotube filled Poly(methyl methacrylate) with unique mechanical, electrical and thermal properties [64]; d, Proposed supramolecules/assembly with design-in dielectric properties. Some of the structures taken from [8, 65].

schematically our perspective for the time scale in the development of nanodielectrics. Other than heterogeneous nanodielectric systems as discussed in this paper, homogeneous nanodielectrics such as ferroelectrics materials with nanosized domains have also many interesting properties for us to explore.

4 CONCLUSIONS

BASED on recent progress in the nanodielectrics field and the electrical power industry needs, we have summarized in the present paper our perspective on the development, applications and time scale in the development of nanodielectrics in the areas shown schematically in Figure 13.

In the short term, incorporation of nanoparticles will make combined progressive electrical, mechanical and thermal improvements in important engineering fields, resulting in many interesting applications such as enamel insulation, electrical grading, EMI shielding, in a cost effective manner.

With proper material/processing developments, nanocomposites based on preformed (mineral or synthetic) anisotropic fillers will have unique anisotropic properties suitable for applications such as electrical discharge resistant machine insulation.

In the long run, with the improving of understanding of dielectric physics at nanoscale, nanostructured dielectric materials/devices could be engineered with many novel dielectric properties and wide applications in energy storage, electro-optic, electro-strictive, and insulating areas.

Fundamental understanding of dielectric properties at the nanoscale level is of great importance in the development of functional nanodielectrics for the electric power industry. More than ever such development needs will require close cooperation between industry and academia. Major original equipment manufacturers for example are already investing tremendous research and development effort and resources to position themselves for the coming challenging decade.

ACKNOWLEDGMENTS

The authors are indebted to the kind help and guidance from the editors, Prof. L.A. Dissado and Prof. J.C. Fothergill. The authors would like to thank Drs. Randall Carter, George Gao, Clive Reed, Nancy Frost, Michael Takemori, Michael Minnick, and Mr. Michael Brown for support and helpful discussions.

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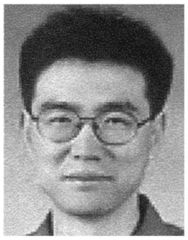
Editor's note: Web sites references are no longer used in IEEE journals because they are not permanent and therefore are not archival. However, since the nanodielectric area is still emerging at the present time some relevant information can only be found on web sites and therefore an exception is made for this paper.



Yang Cao (S'99, M'02) received his B.S. and M.S. degrees in physics from Tongji University in 1992 and 1995, respectively. He completed his Ph.D. in materials science at the University of Connecticut in 2002, working in the area of dielectrics. He is presently with the GE Global Research Center in Niskayuna, NY.

Patricia C Irwin (M'00) earned her Bachelors degree in Chemistry from the College of St. Rose in 1976 and completed graduate work in Chemistry at RPI. She is an electrical insulation engineer with GE Global Research Center in Niskayuna, NY where she is currently involved in the development of new insulation systems for all of GE's various businesses. Prior to that, she was an analytical chemist with GE Power Systems Materials and Processes Laboratory in Schenectady, NY. Ms. Irwin is a member of the IEEE Dielectrics and Electrical Insulation Society.

Karim Younsi (M'91, SM'04) earned his B.S. degree in electrotechnology in 1986 from the University of Science and Technology of Oran and his M. Sc. and Ph.D. in electrical engineering - dielectric materials from Paul Sabatier University in Toulouse, France. Most recently, he was an insulation systems engineer with GE Power Systems in Schenectady. Prior to that, he was an insulation research engineer with GE Canada in Ontario, a research engineer with Ontario Hydro in Toronto, and a research associate with Queen's University in Ontario. In 2003, he joined the Electric Machines and Drives Lab of GE Global Research Center in Schenectady, NY as a dielectrics and insulation systems engineer. He is currently involved in the design and qualification of new insulation systems for a variety of electrical equipments such as medical systems, various motors and generators. Dr. Younsi is a registered member of the Association of Professional Engineers of Ontario in Canada, and a member of the IEEE Dielectrics and Electrical Insulation, Industry Applications and Power Engineering Societies.