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Special Collection: Energy Conversion and Storage in Functional Dielectrics

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ABSTRACT

Functional dielectrics are a group of materials possessing interesting electro-active behaviors, such as variable permittivity, high breakdown strength, ferroelectricity, piezoelectricity, and pyroelectricity, resulting from their capacity to generate and respond to electric fields in a non-linear manner. These properties make them highly desirable for energy conversion and storage applications.

In recent years, there has been extensive research aiming at developing new materials with enhanced performance and novel functionalities. This has resulted in the creation of a diverse form of functional dielectrics, including ceramics, polymers, composites, and thin films, at different atomic, nano, and mesoscales. These functional dielectric materials are commonly used in capacitors, sensors, actuators, nonvolatile memory devices, energy harvesting, and medical instruments, exhibiting intriguing functionalities, phenomena, and manufacturing feasibilities.[1,2] Advanced synthesis and processing techniques, such as solid-state reaction, sol–gel processing, and hydrothermal synthesis, have been developed to tailor the properties of functional dielectrics.[3] Further research on the dielectric materials includes the doping of metal ions, nanofillers, and liquid crystal molecules, as well as structure design of modules to enhance their performance, as they allow for the creation of custom-designed interfaces and layered structures.[4,5] Additionally, theoretical modeling and simulation have been employed to improve the fundamental understanding of the electro-active properties of these materials and to guide the design and exploration of new materials.[6] The development of functional dielectrics is a rapidly evolving field with significant potential for advancing energy conversion and storage technologies, improving the performance of electronic and electromechanical devices, and enabling new applications in several areas, such as renewable energy networks, Internet of Things, and biomedical devices.[7,8]

This Special Topic focuses on energy conversion and storage in functional dielectrics, covering a range of articles in areas of current interest, encompassing synthesis, experimental tests, theoretical calculations, and simulation works. The aim of this Special Topic is to include the interesting research results to inspire future understanding in fundamental science and discovery, as well as applied research for promising devices and systems. We are pleased to see numerous research findings collected in this Special Topic: from inorganic dielectrics and organic polymers to composite materials, from upgrading the conventional preparation methods to the development of new fabrication processes, from modulating the lattice defects or functionalizing the end-group coordination to the design of functional multilayer-structures, from the new understandings of fundamental dielectric mechanisms to the development of practical applications, etc. While, on the basis of the classification of inorganic substances and organic polymers, a series of latest research results on performance improvement, process optimization, in-depth theoretical research, and exploration of dielectric materials assembled in this Special Topic will be introduced.

The initial dielectric-material series has been generally perfected in the past decades of development, but it is still promising to regulate and design the material structure at atomic scale to improve their dielectric properties. For inorganic material systems, element doping is a common and effective approach for substantially improving the

energy conversion and storage performance of dielectrics. The structural heterogeneity induced by various ion-doping provides a promising way to explore new functional-dielectrics. For example, Li's group modified $AgNbO_3$ (ANO)-based lead-free antiferroelectric ceramics with differential A-site and B-site doping, which proved to be an effective route for improving the energy storage density and efficiency of the material.[9] Controlled ion-doping can also be used to construct stepped-wise phase transitions, thereby delaying the polarization process of antiferroelectric ceramics, which can be observed in the results from Zhai's group.[10] Multi-element doping may appreciably increase the opportunity to enhance the dielectric performance of the material. Certainly, the mechanism of the composite action of the doped ions still needs to be further studied; Yang *et al.* and Kang *et al.* offer new insight in this Special Topic.[11,12] In addition to introducing ions to achieve lattice doping, the direct design of composite solid solutions is also an effective means to modulate the dielectric properties of the target material system. Reaney's group designed a hybrid solid solution consisting of B-site Mg-doped Ba(TiMg)O and NaNbO that can withstand pulsed unipolar fields up to 300 kV/cm and achieve considerable recoverable energy density and efficiency.[13] Similar approach of solid-solution design is also quite common in the articles collected in this Special Topic, and some researchers have conducted in-depth studies on the mechanism of defect behavior, polarization regulation, antiferroelectric relaxor-behavior induction, etc., in the hybrid solid solution dielectric systems.[14–17] Nevertheless, the preparation process of conventional solid solutions or ion-doped substances is challenging to produce ultra-thin dielectric films with high quality and homogeneity, which is one of the most important factors which should be considered for integrating and miniaturizing electronic devices in the future. Therefore, it is interesting to implement the rational design to make controllable hybrid structures integrated with functional dielectrics. Hao's group took advantage of the electro-deformation properties of the piezoelectric substrate (PMN-PT) to modulate the Raman characteristics of the ultra-thin two-dimensional black phosphorus in contact with it,[18] while Dong *et al.* used a flexible substrate to regulate the photoluminescence properties of the Ni-doped STO films.[19] Another effective approach is to design and grow superlattice films (such as epitaxial lead zirconate titanate, PZT multilayers, with different components) from the bottom up and regulate their piezoelectric properties by interlayer strain.[20] In addition, as presented in Huang's group, the hybrid structures formed by the combination of polymer coating layer and inorganic dielectric ultra-thin-layer can also produce defect-passivation and other interface optimization effects, which provides more possibilities for the development and application of new inorganic material systems.[21] In addition, the technological barriers existing in the high-quality and low-cost preparation of emerging matrix materials are also being overcome, such as the simple all-room temperature preparation technology of Ag_2Se proposed by Jakhar *et al.*[22]

The functional dielectric material system based on organic polymers is also a very large family with great potential in energy conversion and storage science. Different from the relatively simple inorganic-substances, the design and preparation of variable polymer dielectrics are still emerging research direction. For instance, Shen's group proposed a mechanical stretching strategy for developing the semi-crystalline polymer P(VDF-TrFE-CFE), which is dominated by amorphous phases of disordered entangled chains, into a film with highly aligned molecular chains, greatly improving its thermal diffusion coefficient and thermal conductivity.[23] The preparation strategy of mechanical stretching was also adopted by Xiong's group, and their solution was to disperse small liquid crystal molecules between PVDF molecular chains to create defects, thereby improving the piezoelectric properties of the film.[24] This scheme, also known as "trace nanofiller loading," is an increasingly interesting way to improve the properties of functional-dielectric polymers. In the work done by Zhang's group, the chain packing in the interfacial region of polyetherimide (PEI)-diluted nanocomposites was studied by atomic force microscope-infrared spectroscopy (AFM-IR). It was proved that the flexible bond, that is, the ether group in PEI, plays a vital role in the induction of heterogeneous morphology in the interfacial region and finally makes a major contribution to the great improvement of the dielectric constant of the material.[25] Similar work was done by Shi's group and the nanofiller they used was a $C/SiO_2@TiO_2$ tri-phase nanoparticle with a hybrid nuclear satellite structure.[26] Sardana *et al.* further proposed the use of MXene as a nanofiller. MXene can promote the interface polarization by creating equivalent micro capacitors or osmotic devices, thus giving high charge induction and capture capabilities, and ultimately improve the energy conversion efficiency of nanogenerators based on the principle.[27] In addition, end-group functionalization, composition modifications, and other strategies with chemical bonding reactions also promote the development of polymer–dielectric systems. For example, by combining reversible addition-fragment chain transfer polymerization (RAFT) and additive polymerization techniques, Feng *et al.* created a dithioester-ended-polythiourea-based copolymer dielectric with an extremely high energy density of 10.7 J/cm^3.[28] Qiu *et al.* used ether- and cyano-groups for functionalizing pyrrolidine and then developed it into an electrolyte solution to produce novel high-performance lithium batteries.[29] In addition, the easy preparation of polymers into solutions has greatly increased the possibility of multi-polymer blending to prepare composites with new properties or functions, much higher than that in inorganic systems. Pan's group developed the co-blended PLLA and PDLA into a bio-degradable polymers of stereocomplex crystals poly-(lactic acid), which show high-temperature reliable insulation and mechanical properties, as well as excellent energy density and conversion efficiency.[30] Zhang's group prepared a composite made of polyether imide (PEI) blended with high electron affinity polymer dots (PDs). The ability of PDs to capture free electrons by electrostatic attraction gives this blended composite an extremely high energy density.[31] In this report, the researchers modified the material through ultraviolet irradiation process, and similar post-processing strategies are also very effective in improving the properties of functional dielectric materials, i.e., air-plasma discharging process for advancing PVDF composite systems done by Sasmal *et al.*[32]

In addition to a number of experimental articles on the development of material systems, this topic also includes theoretical research work. Chen's group utilized the finite element method in studying the influence of transverse dimensions on the piezoelectric, dielectric, and pyroelectric properties of $Pb(In_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PIMNT), a new generation of relaxation-ferroelectric materials.[33] Coincidentally, the role of shape factors in the modulation of piezoelectric properties of ZnO nanotubes has also gained new understanding in the theoretical research work of Willatzen's Group.[34] Also, as a traditional functional dielectric material, the negative electrothermal effect of BNBT has been further explored by those researchers in

the Wang's Group in Australia.[35] In addition to the theoretical study of material systems, the exploration of physical models of dielectrics is constantly developing. Barry *et al.* used direct numerical simulation to study the planar capacitance heat transfer characteristics of dielectric liquid under the action of high frequency voltage increase.[36] Yousefian *et al.* developed new lifetime prediction models and metrology methods to help improve the performance of $BaTiO_3$-based multilayer ceramic capacitors.[37] Finally, it is worth noting that Zeggai *et al.* have defined a new metric for evaluating the cooling efficiency of flexible electrocaloric materials, in which dielectric-loss performance becomes the most critical factor.[38] The proposal of the new index will greatly promote the development of functional dielectric materials to more emerging application fields.

With the development of basic material systems and physical mechanism research, the application of functional dielectric substances has been greatly expanded to wireless sensor networks, the Internet of things, implantable biomedical devices, and other emerging fields. However, the accompanying requirements such as higher energy density or conversion efficiency encourage researchers to make more efforts in thin-film formation design, device structure development, operation model optimization, and evaluation index upgrading. Among the articles included in this Special Topic, Mei *et al.* developed an ultra-sensitive H_2O_2 detection module by *in situ* growing ZnO nanoarrays with excellent piezoelectric catalytic properties on flexible gold interfinger electrodes;[39] by means of a purposeful piezoelectric-phase volume fraction modulation strategy, Li's group proposed a vortex-induced underwater piezoelectric-energy harvester based on an optimized PIMNT single crystal macro-fiber composite, which demonstrated considerable power density;[40] Ruan *et al.* prepared polypropylene ferroelectric films with human-skin texture and mechanical structure imitating double-level cellular structure, which creatively promoted the application of ferro-electrets in flexible bioelectronics;[41] Chen *et al.* proposed a two-degree-of-freedom piezoelectric aeroelastic energy harvester for more efficient collection and extraction of wind energy.[42] In addition to new material and device designs, novel strategies has also been made in operating models and schemes. For example, Xiang's group proposed a new high efficiency charging scheme by modulating the product of gas pressure p and electrode spacing d to the valley point of Paschen's law curve by controlling the gas pressure in the cavity, so as to effectively trigger the charging by means of dielectric barrier discharges.[43] With the rapid development and wide application of functional dielectric, we expect that more and more researchers will be involved in more in-depth physical model construction, and further establish, develop, and perfect the working model and performance index of novel dielectric devices. This is of great significance for the development of high-performance dielectric devices that are more in line with the needs of the new generation of information technology and industry.

To summarize, this Special Topic offers readers a valuable opportunity to delve into the latest advancements in dielectric materials and devices. Through this compilation of articles, our objective is to ignite a heightened research enthusiasm and push the potential advantages of functional dielectrics to new frontiers in future applications.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Hong Wang: Writing – review & editing (equal). **Jianhua Hao:** Writing – original draft (equal). **Qing Wang:** Writing – review & editing (equal).

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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