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Ferroelectric polymers with improved performance

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Abstract. This article offers a short overview of the unique properties of PVDF-based electroactive polymers and their applications. Ferroelectric materials are widely used in modern electronics. Relaxor-ferroelectric terpolymers are multifunctional materials with high electrostrictive strain which, in turn, leads to large actuation capability. The terpolymer with clorofluoroethylene shows relaxor-ferroelectric behavior with vanishing hysteresis effect but considerably higher polarization. Therefore, such material is a potential candidate for energy-storage devices (supercapacitors).

Keywords: ferroelectric polymers, polyvinylidene fluoride, polarization, energy conversion, energy storage

Introduction

Polyvinylidene fluoride (PVDF) and its copolymer with trifluoroethylene P(VDF-TrFE) are ferroelectric polymers which exhibit pronounced polarization-versus-field hysteresis and related electromechanical and thermoelectrical response, in particular the piezo- and pyroelectric effect. The terpolymer with clorofluoroethylene P(VDF-TrFE-CFE) shows relaxor-ferroelectric behavior with vanishing hysteresis effect, but considerably higher polarization (Bauer et al. 2004; Klein et al. 2005). Therefore, this terpolymer is a promising candidate for energy-storage devices (supercapacitors) and—given its low elastic modulus—for electromechanical actuators, which use the electrostrictive effect or the Maxwell strain caused by application of a strong DC field.

Since the discovery of piezoelectricity of PVDF in 1969 (Kawai 1969), PVDF and its copolymers have been the subject of numerous structural, dynamic-mechanical, thermal and electrical studies focused on investigating how their preparation-induced microstructure determines their mechanical and electrical behaviour and how this microstructure can be changed by external loads and fields. These studies have been summarized in several review articles and monographs (Furukawa 1989; 1994; 1997; Kepler, Anderson 1992; Kochervinskii 1996; Lovinger 1983; Nalwa 1995).

There have been many successful attempts to use these properties commercially in sensor and actuator applications (piezoelectric transducers, motion detectors, ultrasonic detectors, micro-actuators) and in organic electronics (OFETs) (Bauer, Bauer 2008; Furukawa et al. 2010; Park et al. 2008). An important advantage of ferroelectric polymers in comparison with inorganic ferroelectrics is that they can be processed as flexible free-standing films of only a few micrometers in thickness or as thin submicrometer films on a substrate.

As in inorganic ferroelectrics, the ferroelectricity of PVDF and P(VDF-TrFE) is a property of a polar crystalline phase, but the structure and the elastic properties of the amorphous phase embedding the polar crystallites strongly determines the macroscopic ferroelectric behaviour of the polymers. There exist several polar and non-polar crystalline phases depending on processing conditions and pre-history of the film. Moreover, the intermediate phases between amorphous and crystalline phases must be considered in order to explain the polymers' dynamic-mechanical and electro-mechanical properties. Chemical modification of P(VDF-TrFE) with CFE considerably reduces the crystallite size, leading to a transformation of the ferroelectric-to-paraelectric phase transition into a process that shows features of a strong relaxation. This process determines the high polarization with only little polarization-versus-field hysteresis.

Preparation of ferroelectric polymer films

Films of PVDF and its copolymers can be obtained by solution casting, doctor blading or spin coating from a solution in a polar solvent. With the exception of the copolymer of PVDF with TrFE and the terpolymer, which can both be prepared as (relaxor)-ferroelectric films without further treatment; the other known ferroelectric polymers need subsequent mechanical stretching in order to induce the formation of a ferroelectric phase. This treatment aligns the polymer chains of the amorphous phase in parallel, thus transforming the thermodynamically preferred but nonpolar crystalline α phase into the polar β phase. It is the same polar phase which is formed spontaneously for sterical reasons in the copolymer with TrFE. In order to induce a ferroelectric polarization, the molecular dipoles in the polar phase must afterwards be oriented in an electric field higher than a certain threshold (the coercive field).

During this field treatment, the dipole alignment is stabilized by Coulomb interaction with interface charges accumulated on the phase boundaries between the polar crystallites and their amorphous environment, which are injected from the metal contacts or generated by dissociation of fluorine or impurities (Sessler et al. 1992).

Electro-mechanical response of PVDF and its derivatives in relation to their structure

Up to now, the majority of applications have been based on PVDF-TrFE because of its easy processing as a ferroelectric free-standing thin film or as a ferroelectric film on a substrate and its relatively high piezo- and pyroelectric response, which is only slightly lower than that of the homopolymer PVDF. sThe piezoelectric d_{33} coefficient of PVDF-TrFE is about 20 pC/N, about ten times higher than the piezoelectric response of quartz. However, the thermal stability of PVDF-TrFE ferroelectric polarization is relatively low; it cannot be used at elevated temperatures for longer periods of time. Though there exists a Curie transition at about 100 °C (depending on the TrFE/VDF ratio), the domain orientation decays gradually, because the stabilizing charges flow away or become neutralized at temperatures above the glass transition and because secondary crystallization disturbs the parallel-chain all-trans configuration above 50 °C (Fruebing et al. 2012). The homopolymer is thermally more stable, but in principle exhibits similar mechanisms of polarization decay.

The terpolymer is characterized by a low elastic modulus, which enables a considerable and reversible change of film thickness under mechanical or electrical stress or a deformation of a clamped film, which can be used for the design of micro-actuators. The underlying physical processes are the electrostrictive effect and the Maxwell strain, which is caused by Coulomb attraction between the oppositely charged electrodes.

From normal ferroelectric copolymer into a ferroelectric relaxor

Molecular conformations for PVDF-based polymers are shown in Fig. 1 (Bachmann, Lando 1981). A significant change in various properties, as well as polarization and strain change could be caused by a reversible molecular change from nonpolar to polar forms. Unfortunately, this is not the case for traditional ferroelectric PVDF and copolymers.

It has been shown (Fig. 2) that by introducing defects via high electron irradiation of P(VDF-TrFE) copolymers, the copolymer is converted from a normal ferroelectric to a relaxor ferroelectric. (Bauer 2012; Zhang et al. 1998).

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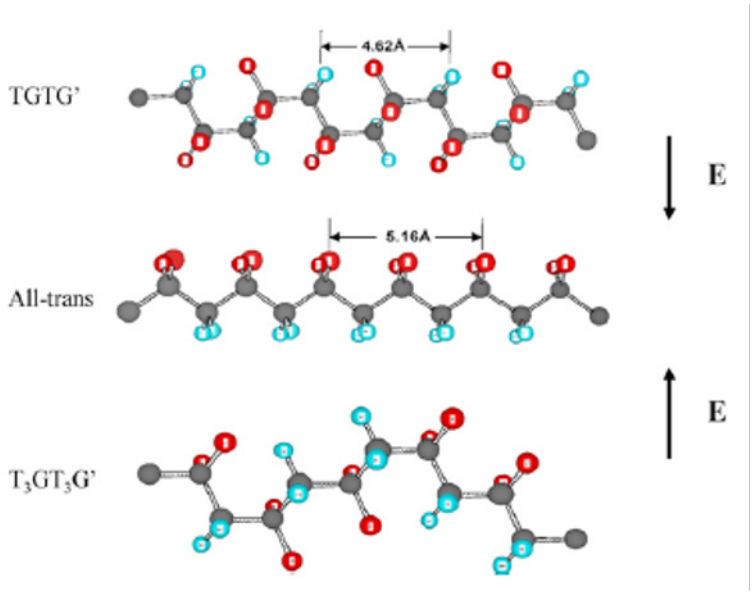

Fig. 1. The conformations of P(VDF-TrFE) copolymer (Bauer 2012)

Here, the polarization practically disappears and the dielectric response shows typical ferroelectric relaxor behavior. The main reason for the observed large electrostrictive strain is the change in conformation from the disordered TG and T_3G to the all-trans conformation.

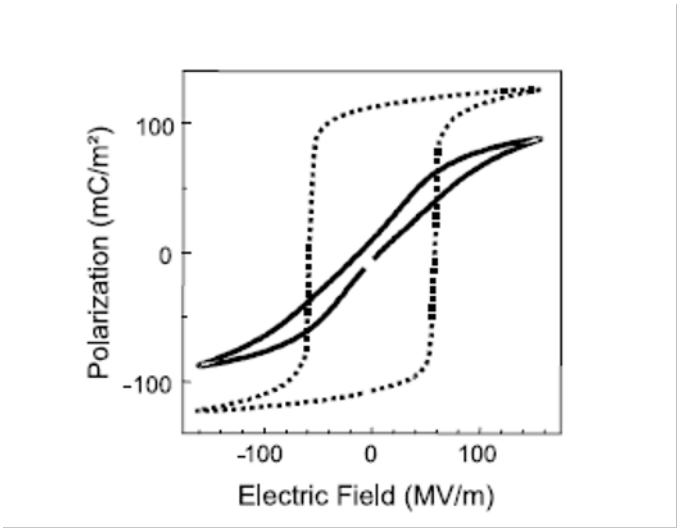

Fig. 2. Influence of defect modifications (Zhang et al. 1998)

Defect modification is another effective way to produce relaxor ferroelectric polymers from P(VDF-co-TrFE) and to modify the Curie transition (Yang, Li et al. 2013). Thus, a new class of ferroelectric polymers, i. e., the terpolymers of P(VDF-TrFE-CFE) and P(VDF-TrFE-CTFE), were synthesized. When the third monomer is introduced into the polymer chain, it interrupts the ferroelectric domains, reducing their size and forcing a conformation change from the all-trans ($T_m \geq 4$) or conformation to the trans-gauche (TG) and T_3G conformations. Such an introduction broadens the ferroelectric transition and reduces the ferroelectric–paraelectric transition temperature (Bachmann, Lando 1981; Bobnar et al. 2003; Huang et al. 2004; Klein et al. 2005; Petchsuk 2003). These relaxor-ferroelectric terpolymers exhibit a room temperature dielectric constant greater than 50 (Bauer, Fousson et al. 2004; Huang et al. 2004; Lu et al. 2006; Xia et al. 2002).

The observed high electrostrictive strain, together with relatively high modulus, makes polymers suitable as structural components in addition to their electroactive functions (actuators and sensors). The amount of monomers added to the P(VDF-TrFE) has a great influence on the strain response and

the polarization hysteresis by changing the spontaneous polarization, the crystallinity, the Young's modulus, the dielectric properties, and the structural conformations (Petchsuk 2003).

Some important applications

Pyroelectric applications

Ferroelectric polymers are a good choice for sensors and energy harvesting devices (Ploss, Domig 1994; Pecora et al. 2012; Setiadi et al. 1999; Whatmore 1986). The most successful ferroelectric polymers for pyroelectric energy harvesting are copolymers and terpolymers, due to their high pyroelectric coefficients (≈ 60 μC m^{-2} K^{-1} for the terpolymer) (Li, Wang 2016).

Electrocaloric refrigeration

Ferroelectrics are preferred electrocaloric materials as they demonstrate large spontaneous electrical polarizations and ferroelectric–paraelectric phase transition, where significant changes in spontaneous electrical polarization take place. They became even more relevant in energy efficient and environmentally-friendly refrigeration (Mischenko et al. 2006; Neese et al. 2008). Here, the idea is to use electrocaloric effect as a technology alternative to conventional vapor-compression, since ferroelectric polymers have much higher breakdown strength in comparison with inorganic materials. Therefore, when a high driving field is applied to the polymers for sizable electrocaloric heat; ferroelectric polymers produce larger Q values. Relaxor ferroelectric polymers show remarkable electrocaloric performance over a wider temperature range than normal ferroelectric polymers (Li et al. 2011; 2012; Peng et al. 2013).

Application in electric energy storage

Polymer-based capacitors demonstrate high energy density, low loss, high reliability, easy processing, and feasibility. Due to the ferroelectricity of (PVDF)-based polymers, they exhibit much higher polarization response under an electric field, in comparison with other linear dielectric polymers for capacitor applications.

The stored and discharged electric energy density for a non-linear dielectric polymer is easily calculated. Fig. 3 shows the polarization hysteresis loop for terpolymer, from which the stored and discharged energy density can be obtained (Bauer et al. 2006). The implementation of defect modifications stabilizes the paraelectric α phase and reduces the remnant polarization (Chu et al. 2006). The discharged energy density of these PVDF-based copolymers is greatly improved over that of the PVDF homopolymer (Chu et al. 2006; Li, Wang 2016).

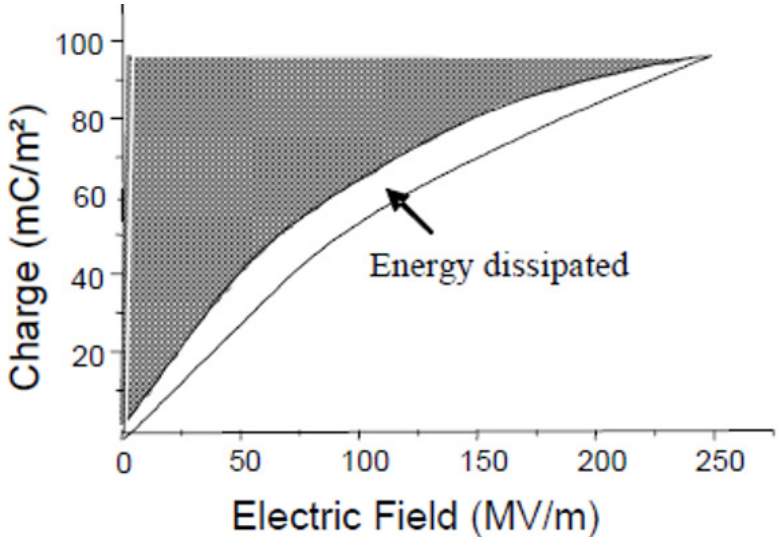

Fig. 3. Determination of discharged/released energy density (shaded area) and dissipation energy (open area) for terpolymer film (Bauer et al. 2006)

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Incorporating a third monomer, such as CTFE and CFE, into P(VDF-TrFE) reduces the ferroelectric domain size and the energy barrier in phase transition, and thereby the normal ferroelectric copolymer is converted to a terpolymer with relaxor ferroelectric behaviour (Bauer et al. 2006; Li, Wang 2016). Relaxor ferroelectricity helps to reduce hysteresis loss and improves the charge–discharge efficiency, while their high dielectric constants promote polarizations.

Actuators

Electromechanical properties of different ferroelectric materials—such as, for example, the maximum strain and the elastic modulus—were compared (Bauer et al. 2007; Cross 1996; Park, Shrout 1997). Terpolymer was found to demonstrate high strain change and high elastic energy density which, in turn, can lead to high motion of a based terpolymer unimorph (Bauer 2012; Zhang et al. 1998). The large electrostrictive strains, which can be induced by an external electric field, are a unique feature of such materials (Bauer et al. 2006; Zhang et al. 2004). These terpolymer properties are very important for the developments of actuators; one of the possible configurations is shown on Fig. 4.

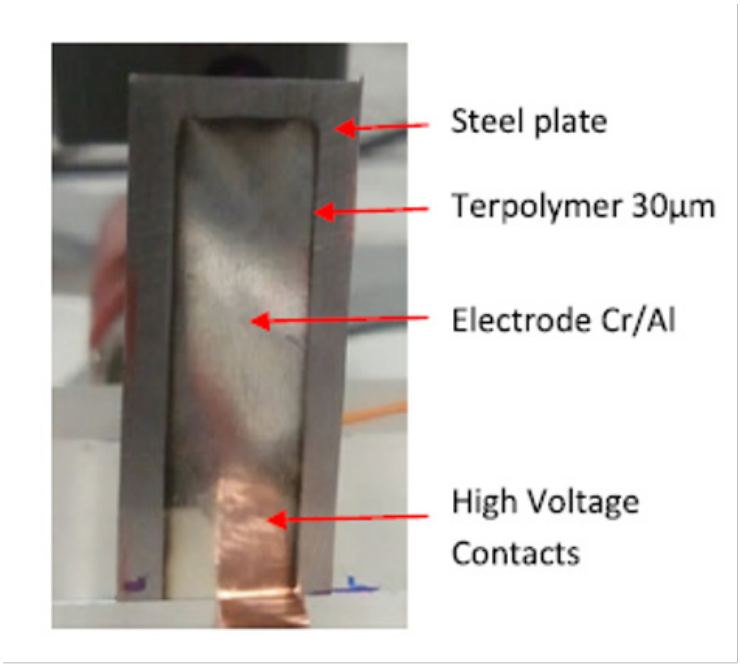

Fig. 4. Actuator configuration for the control of micro pulsed air jet from the shell of a projectile (Bauer 2012; Znang et al. 1998; Xia et al. 2002)

Terpolymers are also promising materials for tactile display applications (Bar-Cohen 2010). It is important to develop a miniature actuator that can meet both the Braille displacement and force requirements while operating at low voltage. Due to the large transverse electrostrictive strains and high elastic modulus, P(VDF-TrFE)-based terpolymers could be successfully used for compact actuation devices (Xia et al. 2002; Zhang et al. 1998). These actuator polymers can be fabricated into thin films (down to 1 μm) enabling low operation voltages for practical commercial devices. Actuators made from P(VDF-TrFE-CFE) and Braille cell design is shown in Fig. 5 (Levard et al. 2012; Bauer 2012). The actuators may also be suitable for a wide range of other applications, including artificial muscles, mechanisms, smart structures, and robotics (Chuc et al. 2008; Rajamani et al. 2008; Ren et al. 2007).

Another device based on electroactive polymer was developed by (Choi et al. 2009) (see Fig. 6). Here, actuators push the optical fluid in the chambers into the lens part, which produces a bending deformation of the elastomer membrane corresponding to the change of focal length (Bauer 2012; Choi et al. 2009).

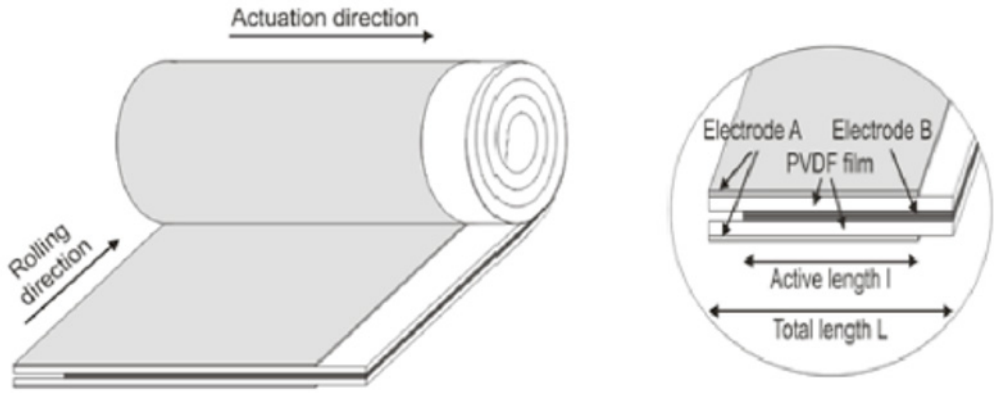

Fig. 5. Actuator design (Levard et al. 2012)

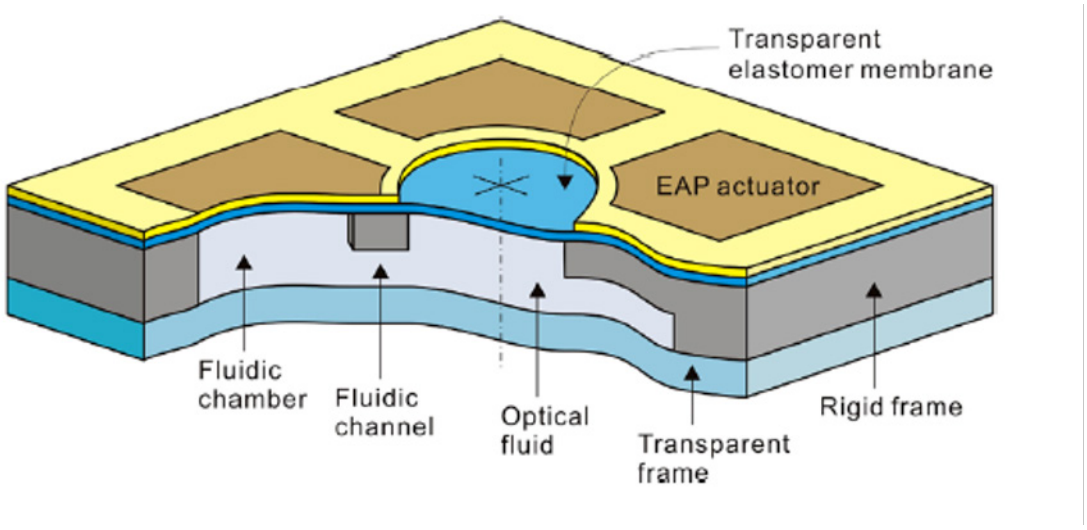

Fig. 6. Scheme of the liquid-filled varifocal lens (Bauer 2012; Choi et al. 2009)

Conclusion

Ferroelectric polymers possess unique physical properties, as well as plenty of advantages of organic materials (processability, flexibility, etc.). Relaxor-ferroelectric terpolymers based on PVDF have become an essential part of advanced electronics, as well as energy storage and conversion devices. Here, some applications of such materials have been presented.

Conflict of Interest

The authors declare that there is no conflict of interest, either existing or potential.

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