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# Impact of structural changes on dielectric and thermal properties of vinylidene fluoride–trifluoroethylene-based terpolymer/copolymer blends

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#### ABSTRACT

We report dielectric and thermal properties of the poly(vinylidene fluoride-trifluoroethylene-chlorofluoroethylene) terpolymer [P(VDF-TrFE-CFE), a member of the relaxor polymer family that exhibits fast response speeds, giant electrostriction, high electric energy density, and large electrocaloric effect] blended with the ferroelectric poly(vinylidene fluoride-trifluoroethylene) copolymer, P(VDF-TrFE). Although the differential scanning calorimetry (DSC) clearly reveals that both components form separate crystalline phases, at low copolymer content blends entirely exhibit a relaxorlike linear dielectric response, since the interfacial couplings to the bulky defects in the terpolymer convert the normal ferroelectric copolymer into a relaxor. On the other hand, dielectric experiments evidence that in blends with 20–50 wt% of P(VDF-TrFE) the ferroelectric and relaxor states coexist. This coexistence is confirmed by DSC results, which further reveal the influence of blending on the terpolymer crystallinity and melting point. At last, the crystallinity data appropriately explain the variation of the dielectric constant in P(VDF-TrFE)/P(VDF-TrFE) blends.

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#### 1. Introduction

Electroactive polymers based on polyvinylidene fluoride, PVDF, and its copolymer with trifluoroethylene, P(VDF-TrFE), are of great interest for a broad range of applications, as they exhibit fast response speeds, and strain levels far above those of traditional piezoceramic materials [1–3]. For example, a giant electrostriction in electron-irradiated P(VDF-TrFE) copolymer [4] and in terpolymers with chlorotrifluoroethylene, P(VDF-TrFE-CTFE) [5,6], or chlorofluoroethylene, P(VDF-TrFE-CFE) [7], has been detected. These systems exhibit a typical relaxor behavior [8], suggesting that electron-irradiation or introduction of CTFE/CFE monomers converts the coherent polarization domain, i.e., all-trans chains, in normal ferroelectric P(VDF-TrFE) into nanopolar regions, thus transforming the material into a relaxor system (a relaxor state is usually described as a network of randomly interacting polar nanoregions which are embedded in a highly polarizable medium) [9,10]. Concomitantly, an ultrahigh strain response in relaxor polymers is generated due to expansion of the polar regions under an external electric field, coupled with a large difference in the lattice strain between polar and nonpolar phases [4].

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Ferroelectric and relaxor polymers have in recent years also been suggested for various advanced applications, as they possess high electric energy density with fast discharge speed [11] and large electrocaloric effect near room temperature [12]. Most of the investigations have, however, focused on either normal ferroelectric polymers or polymers that are completely transformed into a relaxor (e.g., terpolymers or P(VDF-TrFE) copolymer, irradiated with a high dose). Only recently properties of P(VDF-TrFE) copolymer, irradiated with low and moderate doses of high-energy electrons, have been reported - a clear evidence that ferroelectric and relaxor states coexist in the system has been provided on the basis of dielectric and thermal investigations and it has furthermore been shown that such a coexistence strongly influences some materials' properties [13]. Since irradiation also creates some undesirable side effects (crosslinking of polymer chains or formation of radicals, for example), we have developed a polymer system, where similar coexistence of states could be expected - blends of a relaxor terpolymer and ferroelectric copolymer.

We report dielectric and thermal properties of P(VDF–TrFE–CFE) terpolymer/P(VDF–TrFE) copolymer blends. We show that blends entirely exhibit a relaxorlike linear dielectric response at low copolymer content, while in samples with 20–50 wt% of P(VDF–TrFE) the ferroelectric and relaxor states coexist, in terms of the fact that





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**Fig. 1.** Temperature dependences of the real, e', and imaginary, e'', parts of the complex dielectric constant, detected at various frequencies in the pure P(VDF–TrFE–CFE) terpolymer (100:0), its blends with different wt% (5–50) of P(VDF–TrFE) copolymer, and in the pure copolymer (0:100). Due to clarity the *y*-scale of the copolymer sample, which shows a ferroelectric behavior, is different to those of other samples, which exhibit either relaxor behavior or relaxor–ferroelectric coexistence.

samples are composed of the phase separated relaxor terpolymer and ferroelectric copolymer and that their overall response evolves from relaxorlike towards the ferroelectric on increasing the P(VDF– TrFE) content. This coexistence is confirmed by the differential scanning calorimetry, which further reveals the influence of blending on polymers crystallinity and melting points. Moreover, the variation of the dielectric constant in P(VDF–TrFE–CFE)/P(VDF–TrFE) blends is explained in terms of the crystallinity data. A

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