



Contents lists available at ScienceDirect

European Polymer Journal

journal homepage: www.elsevier.com/locate/europolj

Macromolecular Nanotechnology

High-temperature polymer based magnetoelectric nanocomposites



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ARTICLE INFO

Article history:

Received 27 October 2014

Received in revised form 22 December 2014

Accepted 11 January 2015

Available online 17 January 2015

Keywords:

Magnetoelectric

Multiferroic

Polymer

High-temperature

Nanocomposites

ABSTRACT

The use of polymer based magnetoelectric materials for sensing and actuation applications has been the subject of increasing scientific and technological interest. One of the drawbacks to be overcome in this field is to increase the temperature range of application above 100 °C. In this way, a nanocomposite material composed by a mixture of two aromatic diamines, 1,3-bis(2-cyano-3-(3-aminophenoxy)phenoxy)benzene (diamine 2CN) and 1,3-bis(3-aminophenoxy)benzene (diamine 0CN) and CoFe₂O₄ (CFO) nanoparticles was designed, fabricated and successfully tested for high temperature magnetoelectric applications.

Results revealed that CFO nanoparticles are well distributed within the 0CN2CN polymer matrix and that the addition of CFO nanoparticles does not significantly alter the polyimides structure.

The magnetization response of the composite is determined by the CFO nanoparticle content. The piezoelectric response of the 0CN2CN polymer matrix (≈ 11 pC N⁻¹) and the maximum α_{33} value (0.8 mV cm⁻¹ Oe⁻¹) are stable over time and decrease only when the composite is subjected to temperatures above 130 °C. Strategies to further improve the ME response are also discussed.

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1. Introduction

The magnetoelectric (ME) effect defined as the variation of the magnetization by an applied electric field or the variation of the electrical polarization by an applied magnetic field is a scientifically interesting and technological useful phenomenon with a wide range of applications in several

areas such as computer memories, smart sensors, actuators and high frequency microelectronic devices [1–3].

There are very few single-phase ME materials, and most of them show very weak ME coupling at room temperature not allowing their use in most application areas. Composites emerged as a solution to this problem since in composites consisting on magnetostrictive and piezoelectric phases, the ME effect is the result of a product property, i.e., mechanical deformation due to magnetostriction results in a dielectric polarization variation due to the

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piezoelectric effect, allowing high ME effects at room temperature [4,5].

Since the ME coefficient is typically three orders of magnitude higher in ceramic or laminated composites, ME multilayer and ceramic composite materials have attracted particular attention. Laminated composites, however, are difficult to shape and miniaturize as a result of their inherent heterogeneous toughness and the performance of these composites is compromised by the brittleness of the epoxy bonding [4,6,7]. Ceramic composites, also have their restrictions, being limited by deleterious reactions at the interface regions which lead to low electrical resistance and high dielectric losses, hindering sustainable device applications [6].

Magnetolectric composites based on magnetostrictive nanoparticles embedded in a piezoelectric polymer matrix can overcome the above mentioned problems: strain coupling typically does not deteriorate with operation, as the magnetostrictive material is in direct contact with and completely surrounded by the piezoelectric material, have a feasible and scalable production method compatible with industrial requirement and show a flexible structure without large leakage currents [1,6].

Additionally, polymer-based ME composite materials can be fabricated by conventional low-temperature polymer processing into a variety of forms, such as thin sheets or molded shapes, can exhibit improved tailored mechanical properties, flexibility, lightweight, versatility, low cost and in some cases biocompatibility, offering obvious advantages from the point of view of applications [6,8].

Nevertheless in specific areas such as automobile industry, biomedical and machinery used in material processing, most of the polymer-based ME composites do not meet the recent requirement of smart materials working under extreme conditions such as high temperatures [9–12]. Such poor performance at temperatures above 80 °C of polymer based ME materials is related with the fact that most of them are based on poly(vinylidene fluoride) or copolymers, the family of polymers with the largest piezoelectric response [13], undergoing shrinking above this temperature [13,14]. This limitation can be solved by using an amorphous copolyimide containing nitrile groups obtained from a mixture of two aromatic diamines, 1,3-bis-2-cyano-3-(3-aminophenoxy)phenoxybenzene (diamine 2CN) and 1,3-bis(3-aminophenoxy)benzene (diamine OCN), with good piezoelectric performance at temperatures up to 130 °C, as the piezoelectric constituent of ME materials [15].

Such polyimides have received much interest lately due to their excellent thermal, mechanical and dielectric properties [8,16]. It has been further shown that the presence of OCN improves the mechanical properties of the material and that increasing contents of 2CN improved the piezoelectric response [17] to values suitable for applications.

In this way, the aim of the present work is to develop ME composites based on CoFe_2O_4 (CFO)/OCN2CN with suitable ME performance at temperatures above 100 °C.

CFO nanoparticles were chosen as magnetic/magnetostrictive phase due to the large magnetostrictive coefficients, high Curie temperatures and chemical stability [18,19].

2. Materials and methods

Magnetolectric CFO/OCN2CN films were synthesized by the dianhydride ODPa and a mixture of two aromatic diamines, 1,3-bis-2-cyano-3-(3-aminophenoxy)phenoxybenzene (diamine 2CN) and 1,3-bis(3-aminophenoxy)benzene (diamine OCN) with cobalt ferrite magnetic nanoparticles (CFO). The nanoparticles were introduced into a vial containing dimethylacetamide (DMAc), and the vial was placed in an ultrasonic dispersion reactor for 60 min to ensure the uniform dispersion of the CFO on DMAc. Then, a mixture of the two aromatic diamines (50% mole percentage) was added and stirred until the complete dissolution of the diamines. The content of ferrite nanoparticles was 10 weight percentage (wt.%) since this concentration allows suitable nanoparticle dispersion to obtain ME response without affecting composites easy shaping, flexibility and lightweight [1,6,18].

Later, 4,4'-oxydiphthalic anhydride (ODPA) was introduced to the mixture and stirred at room temperature in a dry nitrogen atmosphere resulting in a viscous polyamic acid (PAA) solution containing CFO nanoparticles after 24 h. Finally, the mixture was cast in 150 μm thick films and thermally imidized according to the thermal treatment represented in Fig. 1.

The morphology of the CFO/OCN2CN composites was studied by scanning electron microscopy (SEM) using a Quanta 650 FEI electron microscope with acceleration voltage of 10 kV. Prior to SEM, the samples were coated with gold by magnetron sputtering.

Magnetic hysteresis loops at room temperature were measured using an ADE Technologies vibrating sample magnetometer.

After 60 min of corona poling at 10 kV and at 200 °C in a home-made chamber, following an optimization procedure, the piezoelectric response (d_{33}) of the samples was analyzed with a wide range d_{33} -meter (model 8000, APC Int Ltd.). In order to obtain the out-of-plane ME coefficient, DC and AC magnetic fields were applied along the same direction of the electric polarization of the POCN2CN, i.e., perpendicular to the composite surface. An AC driving magnetic field varying from 0.5 to 1.5 Oe amplitude at

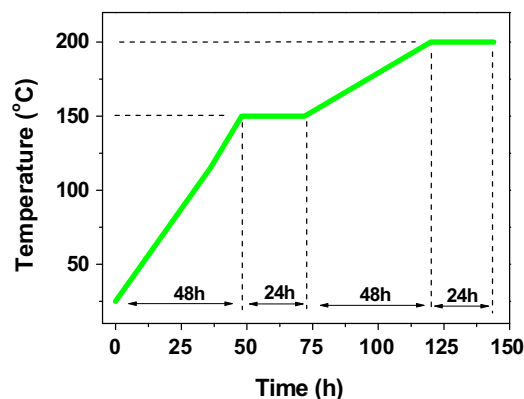


Fig. 1. Thermal treatment used in the preparation of the CFO/POCN2CN ME nanocomposites.

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