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# Silicone dielectric elastomers based on radical crosslinked high molecular weight polydimethylsiloxane co-filled with silica and barium titanate

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**Abstract** A strategy, consisting in the use in tandem of two active fillers in different ratios with complementary effects: silica mainly as a reinforcing agent and barium titanate as dielectric permittivity enhancer, was proposed to optimize the electromechanical properties of the silicone elastomers. A high molecular mass polydimethylsiloxane-$\alpha,\omega$-diol ($M_w = 642000$ g mol$^{-1}$) was used as a matrix to prepare silicone composites which further were processed as films and crosslinked at high temperature. The morphological, thermal, and moisture behaviors of the films were studied by adequate techniques. The mechanical properties were estimated on the basis of normal and cyclic stress–strain curves. Dielectric spectra were recorded in the frequency range of 1–$10^6$ Hz at normal temperature. The voltages generated by a mechanical impulses created by falling of a metal ball of 7.1 g from a height of one hundred millimeters on the surface of the films placed between two electrodes ranged between 28.4 and 157.3 V mm$^{-1}$.

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## Introduction

Energy-harvesting technologies aiming the conversion of different energy forms (light, thermal, mechanical, etc.) [1] from the natural sources such as wind, waves, or animal movements into electrical energy are attracting a high interest in the scientific community especially in the last 20 years [2, 3]. One way to convert the mechanical energy into electrical one is to use high reversible deformability of the dielectric elastomers. Among this class of materials, silicones attract great interest because of their high elasticity and weather resistance [4–8]. A drawback of silicones is their low dielectric permittivity (in the range of 2.5–3.0); many studies thus aim to improve this parameter to enable them maintain good mechanical properties. The unique molecular structure and chemistry of the silicone polymers permit their formulations to be able to meet the requirements of a specific application. Different strategies have been used to improve the dielectric properties [9], but the most promising one is the incorporation of filler particles with high dielectric permittivity, such as ceramic fillers (e.g., barium titanate, titanium dioxide, calcium copper titanium oxide, lead zirconate, etc.) [10–14]. The dielectric relaxation of the crystallites embedded into the polymer matrices was studied [15, 16], and the principal role of the interface states on the border polymer-crystallites was established in defining the corresponding dielectric features. Barium titanate, a ferroelectric crystal with high dielectric strength, is widely used for this purpose [17–20]. The effects of barium titanate ($BaTiO_3$) nanoparticles on electric and mechanical properties are extensively studied, and it was found that the dielectric constant of nanocomposites significantly increases with the increasing $BaTiO_3$ concentration, whereas volume resistivity decreases continuously [21]. However, while the