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## **ACCEPTED MANUSCRIPT**

### Superior dielectric properties in Na $_{0.35\%}$ Ba $_{99.65\%}$ Ti $_{99.65\%}$ Nb $_{0.35\%}$ O $_3$ /PVDF composites

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#### **Abstract**

In this paper, the recently developed environmentally friendly lead-free ferroelectric Na<sub>0.35%</sub>Ba<sub>99.65%</sub>Ti<sub>99.65%</sub>Nb<sub>0.35%</sub>O<sub>3</sub> (NNBT) was firstly used as filler to synthesize ceramic-polymer composites using polyvinylidene fluoride (PVDF) as martix. The composites PVDF-xNNBT with different volume fractions of x = 0, 0.1, 0.2, 0.3, 0.4, and 0.5, were prepared via solution mixing and hot pressing method. Dielectric properties were investigated in the temperature range from room temperature to 250 °C and frequency range of  $10^2$  to  $10^6$  Hz. All the investigated composites show high dielectric constant (>100) and low loss tangent (<5%). The best dielectric properties were found in PVDF-0.5NNBT, which shows weakly frequency and temperature dependent dielectric constant of 220 and low loss tangent of 0.037 at 1 kHz. Interfacial relaxation arising from the ceramic-polymer interfaces was suggested to be the main reason of the superior dielectric properties.

Key words: PVDF polymer; ferroelectric ceramic; composite; dielectric constants; dielectric loss

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#### 1. Introduction

The ceramic-polymer composites (CPCs), prepared by dispersing ceramic powders into polymer matrix, combine the merits of ceramics and polymers and thus hold tremendous promise for a wide range of applications, especially for electric device miniaturization and energy storage [1]. High dielectric constant is a key requisite for these applications. However, dielectric constant of most CPCs is less than 50 even with the ceramic fraction reaching 50% [2]. To overcome this shortcoming, colossal dielectric constant ( $\mathcal{E}' > 10^4$ ) ceramics, such as

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