

Novel Polymer-Ceramic Nanocomposite Based on New Concepts for Embedded Capacitor Application

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ABSTRACT: Novel polymer-ceramic nanocomposite based on new concepts was developed in this work. A high dielectric constant (ϵ_r above 60) was obtained by using the combination of BTO/nickel nanoparticle as the ceramic filler. On the other hand, using Pc-coated BTO, the dielectric constant of nanoparticle was above 80 at 1 MHz.

INTRODUCTION: Recently, with the rapid development of advanced IT technology, finer function and further miniaturization of electronics are required. In order to meet these requirements, polymer-ceramic composites have been of great interest as embedded capacitor materials because they combine the processability of polymers with the high dielectric constant of ceramics. The following two technologies are reported in this work to achieve such nanocomposites.

Phthalocyanine-coat BTO (Pc-coat BTO) was also proved to be workable to increase the dielectric constant of nanocomposite. By heating the mixture of BTO with 1,3,5-tri(3,4-dicyanophenoxy)benzene gradually to 300 °C, a blue modified BTO particles coated by phthalocyanine (Pc-coat BTO) was obtained. Its dielectric constant was detected to above 80 at 1 MHz from such modified BTO/Polymer composite.

Metal nano-particles (Ni) was chosen as an additive to increase the dielectric constant. A high dielectric constant (ϵ_r above 60) was obtained by using the combination of BTO/nickel nano-particles as the ceramic filler. Here, two kinds of nickel nanoparticle were studied with the diameter of 0.4 and 0.15 μm , respectively. It was found that the size of Ni nano-particle also played a great role in their electronic property. With the smaller particle selected, the higher dielectric constant would be detected. However, from the SEM patterns it demonstrated that the sufficient dispersion of BTO and Ni particle was very important to achieve satisfactory samples.

EXPERIMENT: Typical Pc-coated BTO was successful to be prepared in a scale of 500 g. Thus to a solution of 6-cyano in 500 mL DMAc was added 500g 500 nm BTO. The slurry was stirred for two days, and then the solvent was removed by rotary evaporation. The solid mixture was grinded and dried at 40 °C overnight. Then it was introduced to a plate model enwrapped with copper foil in a hot-pressure. The temperature was gradually increased to 300 °C and kept for 1 hour. A blue solid was formed and crashed into fine powder.

Polymer-ceramic composites were developed using commercial epoxy resin with two inorganic fillers: BaTiO₃ (BTO, 0.9 μm) and nickel. The average particle radius of Ni nano-particles are 0.4 and 0.15 μm , respectively. To epoxy resin was added ceramic fillers with certain volume ratio, a homogeneous paste with good particle dispersion was

obtained after mixing for 10 min using hybrid mixer. After film casting, the samples were prebaked at 130 °C for 30 min, then cured for 1 h at the temperature of 180 °C under pressing.

RESULTS & DISCUSSION: (1) *Pc-coat BTO system* By using BTO coated by phthalocyanine instead of commercial BTO, the electronic property of nanocomposite changed greatly, especially their dielectric constant increased a lot (above 80). Details see Table 1. The BTO surface modifications by phthalocyanine were considered to improve the affinity between BTO nanoparticles and matrix polymers (polyamide/BMI, or epoxy resin). Moreover, abundant nitrile groups were also expected to contribute to the dielectric constant due to its high molar polarization (11.0 cm³/mol) and low molar volume (19.5 cm³/mol).

(2) *Ceramic bimodal system* We selected BTO bimodal system (0.9 and 0.3 μm) and BTO/ Ni bimodal system (0.3 and 0.4 μm) as ceramic filler to prepare nanocomposite respectively. From the results demonstrated in Fig. 1, it showed that with the increasing of volume ratio of ceramic part in the composite up to 50%, its dielectric constant increased steadily. After then the tendency became dull because of some void in their cross section confirmed by SEM. When the ratio of ceramics above 80%, the sample obtained was too brittle to carry out any further characterization. And also the result indicated that bimodal system seemed superior to that of unimodal one.

Table 1. The electric property of Polyamide/Pc-coat BTO nanocomposites

BTO vol%	Thickness μm	Electric Property	Frequency			
			1K	10K	100K	1M
60	119	ϵ_r	53.2	52.4	50.3	47.9
		$\tan\delta$		0.262	0.058	0.022
70	39	ϵ_r	92.9	88.8	85.6	82.3
		$\tan\delta$	0.464	0.082	0.029	0.057

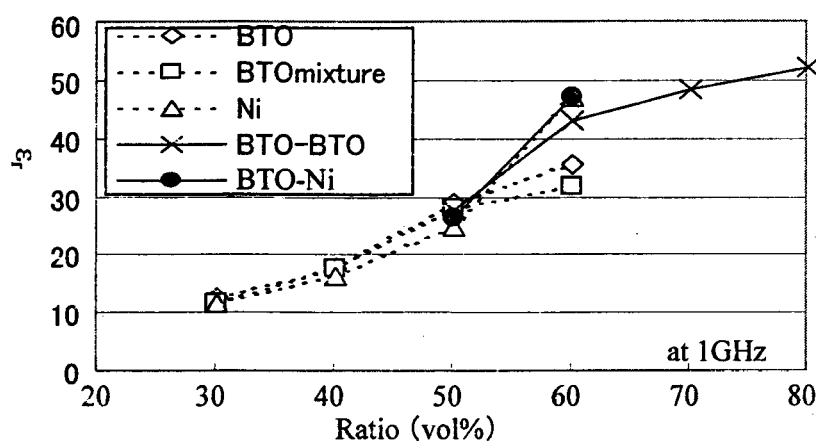


Fig.1 The relationship between ϵ_r and volume ratio of ceramic filler.