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CNTs-added PMNT/PDMS flexible piezoelectric nanocomposite for energy harvesting application

Napatporn Promsawat[1], Methee Promsawat[2], Phatthanapong Janphuang[3], Zhenhua Luo[4], Steve Beeby[5], Catleya Rojviriya[3], Phakkhananan Pakawanit[3] and Soodkhet Pojprapai[1]

[1]School of Ceramic Engineering, Institute of Engineering, Suranaree University of Technology, Nakhon Ratchasima,30000, Thailand

[2]Department of Materials Science and Technology, Faculty of Science, Prince of Songkla University, Songkhla, 90112, Thailand

[3]Synchrotron Light Research Institute, 111 University Avenue, Muang District, Nakhon Ratchasima, 30000, Thailand

[4]School of Water, Energy and Environment, Cranfield University, Cranfield MK43 0AL, UK

[5]Electronics and Computer Science, University of Southampton, SO17 1BJ, UK

*Corresponding author. Email: soodkhet@g.sut.ac.th (S. Pojprapai)

Abstract

The flexible piezoelectric nanocomposites based on lead magnesium niobate titanate [$Pb(Mg_{1/3}Nb_{2/3})_{0.65}Ti_{0.35}O_3$; PMNT] particles in polydimethylsiloxane (PDMS) matrix were fabricated and characterized. PMNT powders are synthesized using the columbite precursor method. PMNT/PDMS flexible nanocomposites are then prepared by spin casting technique, where a small amount of carbon nanotubes (CNTs) is added into the PMNT/PDMS composite to enhance cross-links between PMNT particles and PDMS matrix. The phase and microstructure of the nanocomposite are investigated by using X-ray diffraction and scanning electron microscope (SEM). The electromechanical behavior is evaluated by using an

autonomous pneumatic actuator. The flexible composite, occupying approximately 300 mm^2, is capable of generating an open-circuit voltage (V_{oc}) of 2.83±0.24 V and a short-circuit current (I_{sc}) signal of 0.33±0.01 μA across 10 Ω resistor under mechanical load of 300 N. The generated electrical charges are 29026 pC. The relative dielectric constant is measured at 10 kHz and found to be 6.76±1.15. The piezoelectric PMNT/PDMS composite can be potentially used in a variety of applications such as wearable sensors, actuators, and energy harvesting for converting kinetic energy into useful electrical energy.

Keywords: PMNT, polydimethysiloxane (PDMS), carbon nanotube, nanocomposite, energy harvesting

1. Introduction

Nowadays, renewable energy such as solar, vibrations, electromagnetic wave and thermal energy are widely used in technological applications. These ambient energies have been received much attention from many researchers for applications in the energy conversion technology. The ambient energy sources can be converted to electrical energy via energy conversion processes, i.e. electromagnetic induction [1-2], triboelectric generator [3], electrostatic transduction [4], or using energy conversion materials, e.g., electroactive ferroelectret [5] and piezoelectric materials [6]. Piezoelectric materials are one of the energy conversion materials that can convert kinetic energy from mechanical vibrations to electrical energy via direct piezoelectric effect. Vice versa, they can convert electrical energy to mechanical strain, which is called the converse piezoelectric effect. Using the direct effect, piezoelectric materials can be used to harvest kinetic energy generated from human activities (e.g. walking and running). To date Pb-based piezoelectric materials are the most used in applications due to their high electromechanical coupling factor (*k*) and stimulating at resonance frequencies could realize an energy conversion efficiency above 80% [7]. Of these,

lead magnesium niobate titanate (PMNT) materials is of great interest due to their high electromechanical coupling coefficient which make them as one of promising solution for piezo-sensors and actuators applications [8-9]. The composition of PMNT are typically $Pb(Mg_{1/3}Nb_{2/3})_{1-x}Ti_xO_3$ and it is well-known as relaxor-based ferroelectric material. The composition with x = 0.30-0.35 is at morphotropic phase boundary (MPB) region demonstrating best piezoelectric properties [10]. However, the ceramic form of these PMNT materials is brittle and heavy, making them unsuitable for flexible devices. This disadvantage can be overcome by flexible piezoelectric ceramic-polymer composite [11-12]. The particle size of ceramic powder in the composite is one of the main factors that could affect the piezoelectric properties of the composite. Nevertheless, there were a few works studying the effect of the size of piezoelectric particles on piezoelectric properties of the flexible piezoelectric composites. For example, the work studied by Lee *et al*. reported that the dielectric and piezoelectric constants of the piezoelectric composite with $PbTiO_3$ as the piezoelectric phase increased as the size of the piezoelectric particle increased [13]. Although most polymers exhibit poor piezoelectric properties and dielectric constant, their high flexibility, and forming and shaping abilities make them suitable to be a matrix phase for piezoelectric composites [14-15]. To fabricate piezoelectric ceramic-polymer composite, Polydimethylsiloxane (PDMS) was frequently used as a polymeric matrix due to its flexibility, biocompatibility, and optical transparency. Importantly, it is cost-effective, low toxicity, low permeability to water, high oxidative and high thermal stability [16-18]. Recently, Park *et al*. developed a flexible nanocomposite generator based on PZT particles distributing in a PDMS matrix with added CNTs [19]. The output voltage and current signals of the device were 10 V and 1.3 μA. Furthermore, $NaNbO_3$ nanowires-PDMS composites with output voltage and current of 3.2 V and 72 nA have been synthesized by Jung *et al*. [20]. Chen *et al.* fabricated piezoelectric nanogenerators based on PZT nanofibers which generated the output voltage and

power under periodic stress application of 1.63 V and 0.03 μW, respectively [21]. Jeong *et al.* successfully produced lead-free flexible nanocomposite generators by a simple spin-casting method. The output voltage and current signals, which were generated during repeated bending and unbending motions with a maximum displacement of 5 mm at a rate of 0.2 m s^{-1}, were 12 V and 1.2 μA, respectively [22]. To date, based on our knowledge, only few studies have been reported on flexible piezoelectric composite based on PMNT in PDMS matrix [23-25].

In this work, flexible piezoelectric composites based on $Pb(Mg_{1/3}Nb_{2/3})_{0.65}Ti_{0.35}O_3$ (PMNT) particles dispersed in PDMS polymeric matrix were fabricated. 1wt% CNTs was added into the composite in order to improve the electrical conductivity path between micron-sized piezoelectric particles while maintaining the domain switching during poling process. Physical and electromechanical properties such as phase, microstructure, dielectric properties, and power output of the fabricated composite were then characterized in order to obtain more detailed description of the piezoelectric nanocomposite.

2. Experimental Details

$Pb(Mg_{1/3}Nb_{2/3})_{0.65}Ti_{0.35}O_3$ (PMNT) powders employed as the dispersed phase in this research were prepared by the columbite precursor method from PbO, MgO, Nb_2O_5 and TiO_2 precursors [25]. Firstly, MgO and Nb_2O_5 powders were mixed and then calcined at 900 °C for 4 hr to produce columbite ($MgNb_2O_6$) powders. The $MgNb_2O_6$ powder was then mixed with PbO and TiO_2 powders and calcined at 850 °C for 2 h to produce PMNT powders. Polydimethysiloxane (PDMS) Sylgard 184 from Dow Corning was used as the matrix phase. PDMS elastomer was mixed with a curing agent using the weight ratio between the elastomer and the curing agent of 10:1. The mixture was exposed to thermal curing in order to harden PDMS. 12 wt% PMNT particles were dispersed in the PDMS elastomer in a beaker. After that, 1 wt% CNTs (NANOCYL^{TM} NC7000) was added into the mixture and desiccated under vacuum for 20-30 min to remove air bubbles formed during the mixing process. The layered composites, as

shown in Fig. 1(a), were fabricated by the following steps; (i) spinning PDMS polymer onto a glass plate and then curing at 85 °C for 1 h in an oven (ii) spinning CNTs-added PMNT/PDMS mixture onto the PDMS layer and then curing at 85 °C for 1 h in an oven and (iii) spinning PDMS polymer onto the mixture layer and then curing at 85 °C for 24 h in an oven. After these steps, a flexible piezoelectric composite with a thickness in the range of 200-300 μm was achieved as illustrated in Fig. 1(b). Both sides of the layered composite were etched by oxygen plasma before electrodes were deposited. The silver electrodes were sputtered on plastic substrates (200 μm in thickness) and attached onto the etched surfaces using unsolidified PDMS as a binder. The composite electrodes were cured in an oven at 85 °C for 1 h. Each composite sample was poled under an applied DC electric field of 2.5 kV/mm at 80 °C for 15 h in silicon oil bath. An electrical poling process was required to align the dipole moment to the same direction and to enhance piezoelectric properties of flexible nanocomposite devices. The phases of samples were identified by X-ray diffraction (XRD) technique (Bruker D8 Advance) with Cu K_α radiation wavelength of 1.54 Å. Frequency dependence of a dielectric constant (ε_r) and loss tangent ($\tan\delta$) was measured using a LCR meter (GW INSTEK LCR-821). Microstructure was observed by a field emission scanning electron microscope (FE-SEM, JEOL JSM-7800F). Chemical composition was analyzed by an energy dispersive X- ray spectroscopy (EDX). The internal distribution of the dispersed and matrix phases was investigated using synchrotron X-ray computed tomography. The synchrotron X-ray beam with energy range of 2-18 keV (white beam) was scanned on the sample. The Electromechanical behavior of the composites was investigated using an in-house developed autonomous pneumatic actuator. The output voltage and current under mechanical load were measured using an oscilloscope (Tektronix MSO 3034) and a multi-meter (Agilent 34461A).

3. Results and Discussion

X-ray diffraction patterns of PMNT powder, PDMS polymer, and PMNT/PDMS nanocomposite are shown in Fig. 2. It is obvious that the XRD pattern of the PMNT powder was well-matched with the ICSD file No. 157487 of $Pb(Mg_{1/3}Nb_{2/3})_{0.65}Ti_{0.35}O_3$ compound with tetragonal P4*mm* space group. No secondary phase was observed on the pattern. The XRD pattern of the PDMS indicated an amorphous phase of the sample. A broad peak at $2\theta \approx 12°$ was found, which is consistent with the previous observation of Niu *et al*. [27]. In addition, for the composite, unknown peaks were observed at $2\theta \approx 28°$. The unknown peaks were believed to be possessed by $Pb_{1.83}Nb_{1.71}Mg_{0.29}O_{6.39}$ pyrochlore phase (JCPDS file No. 33-0769). The splitting of (002) and (200) peaks at $2\theta \approx 45°$ was also presented. None of CNTs pattern was observed in the composite. Since only 1 wt% CNTs was added, it is likely that such small amount was below the detection limit of the XRD apparatus.

SEM images of a cross-sectional view of a composite is shown in Fig. 3(a). The layer of the CNTs-added PMNT/PDMS material with 203.58±1.96 μm of thickness was observed in the middle region of the composite sandwiched by the PDMS layers. It can be seen from Fig. 3(b) that the particle size of PMNT phase was about 3 μm. It was clear that CNTs bridged PMNT particles to the matrix, as indicated by the arrow in Fig. 3(b) and (c). This contributed to an improvement of a transportation of charge carriers between the dispersed phase and the matrix phase while maintaining domain switching in a poling process. This could be confirmed by a result observed by Gong *et al*., that the applied field amplitude used in poling CNTs-added composite was lower than the composition without CNTs [28]. A synchrotron X-ray image of CNTs added PMNT/PDMS composite is shown in Fig. 3(d). The different contrast is depended on the level of X-ray absorption of each phase composed in the composite. As shown in the figure, the dark and bright regions represented the locations of the PMNT and PDMS phases, respectively. The brightest region indicated the position at which voids or porosities were located, resulting in a reduction of X-ray absorption at those positions of materials. By using

the synchrotron X-ray imaging technique, the dispersion of PMNT particles in PDMS polymer matrix can be determined. It was also found from the figure that the average size of the dark region was about 30 μm. This indicated the agglomeration of PMNT particles. Although the particle size of PMNT was small, the agglomeration of the PMNT particles and the bridging between the dispersed and the matrix phases of CNTs could contribute to a good piezoelectric properties of the composite. Energy dispersive X-ray spectroscopy (EDX) of CNTs-added PMNT/PDMS nanocomposite was shown in Fig. 4(a) and (b). The results clearly showed the present of Pb, Mg, Nb, Ti and O peak in the PMNT particles and showed C peak representing local CNTs.

Frequency dependences of relative permittivity (ε_r) and dielectric loss ($\tan\delta$) measured at room temperature of CNTs-added PMNT/PDMS nanocomposite is shown in Fig. 5. The result showed that ε_r of the composite suddenly decreased while $\tan\delta$ increased when frequency increased from 1 kHz to 10 kHz and it decreases when the frequency is greater than 10 kHz. The decreasing tendency with frequency of dielectric constant could be caused by an interfacial relaxation. The interfacial polarization arise as a result of difference in conducting phases, but was interrupted at grain boundaries due to lower conductivity. Generally, in polycrystalline materials, the grain exhibits semi-conducting behavior while the grain boundary is insulator. At higher frequency, the charge carriers will no longer be able to rotate sufficiently rapidly, so their oscillation will be laid behind the field resulting in a decrease of dielectric permittivity [29]. The dielectric losses at low frequencies are much higher than those occurring at high frequencies. However, the permittivity of the composite is still above 5. It was found in a previous result observed by Du *et al*. that a monolithic PDMS and PDMS based materials showed dielectric constant in the range of 2.32-2.40 [30]. Furthermore, Romasanta *et al.* [31] reported that the nanocomposite containing 1.0 wt% of CNT influenced to electrical insulator behavior with a permittivity constant of 1.5 times. It is ascribed to the motion of free charge

carriers due to the formation of a continuous conductivity pathway throughout the medium between CNTs. Therefore, it could imply that an addition of PMNT and CNTs improves dielectric properties of PDMS polymers.

The electrical characterization of PMNT/PDMS nanocomposites with an area of 30 mm × 10 mm is illustrated in Fig. 6(a). The output voltage and generated current under an applied mechanical load of 300 N were about of 2.83±0.24 V and 0.33±0.01 μA. The calculated output power of this device is 0.934 μW at load resistance of 10 ohm. As compared to the piezoelectric generators in literature [20,32,33], it was found that the output power of our nanocomposite was lower than that of the flexible PZT-based composite, which is equal to 13 μW, but higher than those of the $BaTiO_3$ thin film and $NaNbO_3$ nanowires-PDMS composite piezoelectric generators, which are equal to 0.026 and 0.23 μW. Fig. 6(b) showed the output current versus time of the composite. The charge generated by the composite can be determined by integrating the current over the time. Using an applied force of 300 N, the average generated charge was equal to 29026 pC. By dividing the charge with the force, the piezoelectric constant (d_{33}) of the composite was calculated which was about 97 pC/N. Moreover, with the permittivity measured at 1 kHz of 10.5, the piezoelectric voltage constant (g_{33}) was calculated to be about 1.04 V·m/N. As compared to PZT/PDMS composites prepared in the previous works by Babu *et al*. [33] and Sharma *et al*. [34] at the equivalent amount of the piezoelectric elements (~ 2 vol%), it was found that our CNTs-added PMNT/PDMS nanocomposite showed higher d_{33} and g_{33}. The d_{33} and g_{33} values of the PZT/PDMS composites were below 40 pC/N and 50 mV·m/N, respectively. The greater of piezoelectric properties of CNTs-added PMNT/PDMS nanocomposite is possibly due to two contributions; 1) low coercive field (E_c ~ 7 kV/cm) with large remanent polarization (P_r ~ 31 μC/cm^2) of the PMNT material [26] and 2) bridging a PMNT particle to the neighboring particles and PMNT particles to PDMS matrix by added

CNTs. These could allow most dipole moments of PMNT phase to switch their directions to the direction of applied field in the poling process.

4. Conclusion

CNTs-added PMNT/PDMS flexible nanocomposites have been successfully fabricated. The electrode was prepared by sputtering silver onto plastic substrate. Both sides of the composite were etched by oxygen plasma before electrodes were attached. XRD pattern of the composite indicated PMNT phase and PDMS phase. PMNT particles were dispersed around in PDMS polymer matrix and bridged the neighboring particles and PDMS polymer by CNTs. Dielectric permittivity and loss tangent of the composite almost decreased with an increase in frequency. The voltage and current output were found to be 2.83±0.24 V and 0.33±0.01 μA, equivalent to 0.934 μW. Under an applied force of 300 N, the composite generated the electrical charge of 29,026 pC. The calculated piezoelectric and voltage constants were 97 pC/N and 1.04 V·m/N, respectively. The use of CNTs as a bridging conductive material could facilitate a poling performance, thereby improving piezoelectric properties of CNTs-added PMNT/PDMS nanocomposites which are suitable for flexible electronic devices and sensor applications.

Acknowledgments

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Figure captions

Fig. 1 (a) Schematic of layered CNTs-added PMNT/PDMS nanocomposite and (b) showing high flexibility of the composite.

Fig. 2 XRD patterns of monolithic PMNT particles and PDMS polymer, and a CNTs-added PMNT/PDMS nanocomposite.

Fig. 3 SEM images of microstructures of (a) cross-section (b) an agglomerated PMNT particles (c) CNTs bridging PMNT particles and PDMS polymer of a layered CNTs-added PMNT/PDMS nanocomposite and (d) tomographic of the particles distributed in the polymer phase using synchrotron X-ray tomography.

Fig. 4 Energy dispersive X-ray spectra of (a) CNTs phase and (b) PMNT phase indicated by arrows in the CNTs-added PMNT/PDMS nanocomposite.

Fig. 5 Frequency dependence of dielectric constant (black line) and dielectric loss (red line) of CNTs-added PMNT/PDMS nanocomposites.

Fig. 6 Cycling output (a) voltage and (b) current under an applied mechanical force of 300 N of CNTs-added PMNT/PDMS nanocomposite.

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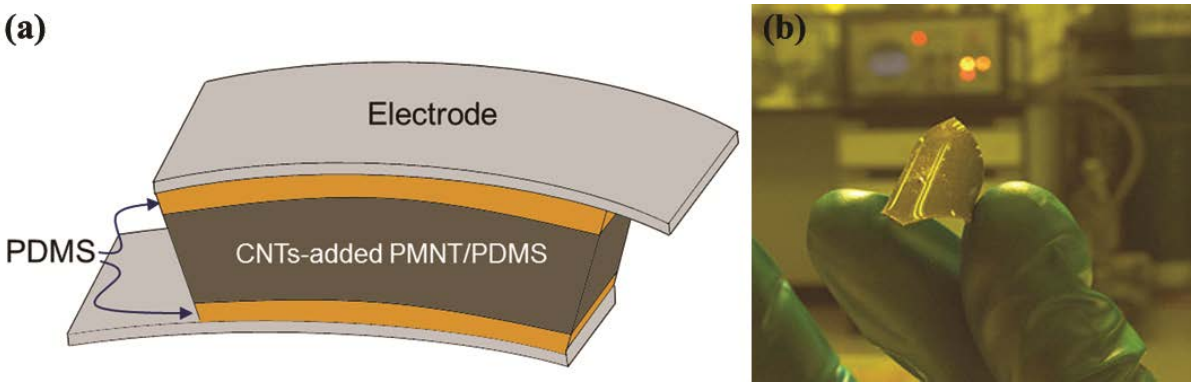
2 Fig. 1



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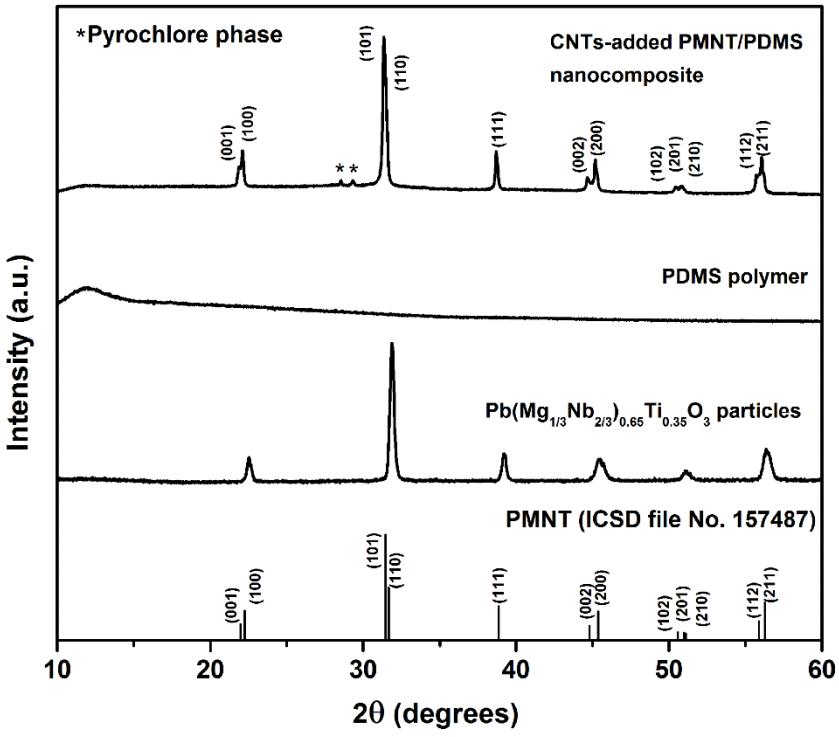
6 Fig. 2



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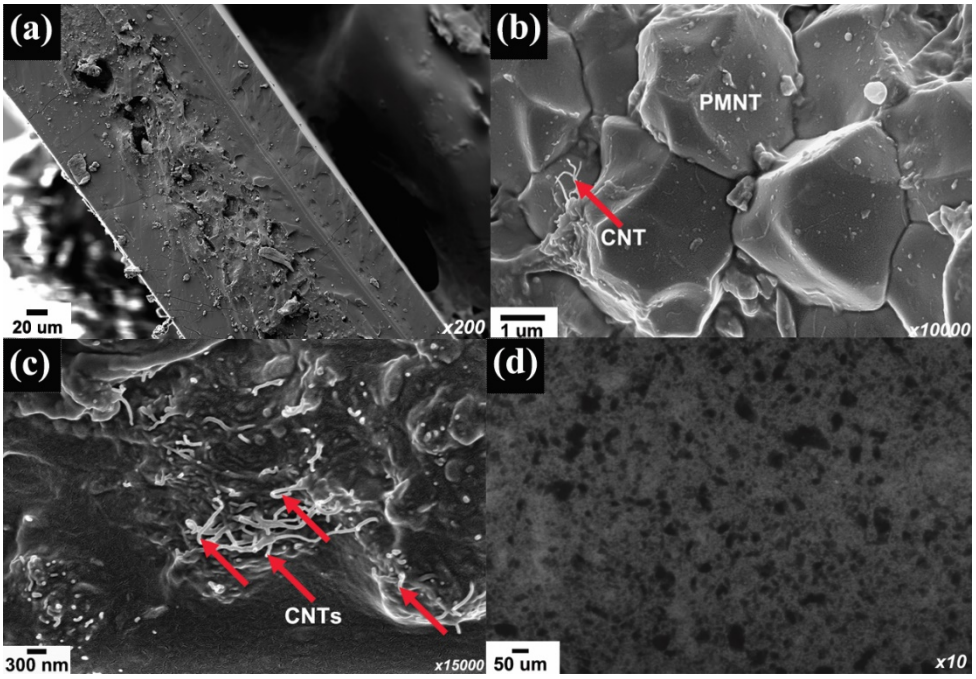
2 Fig. 3



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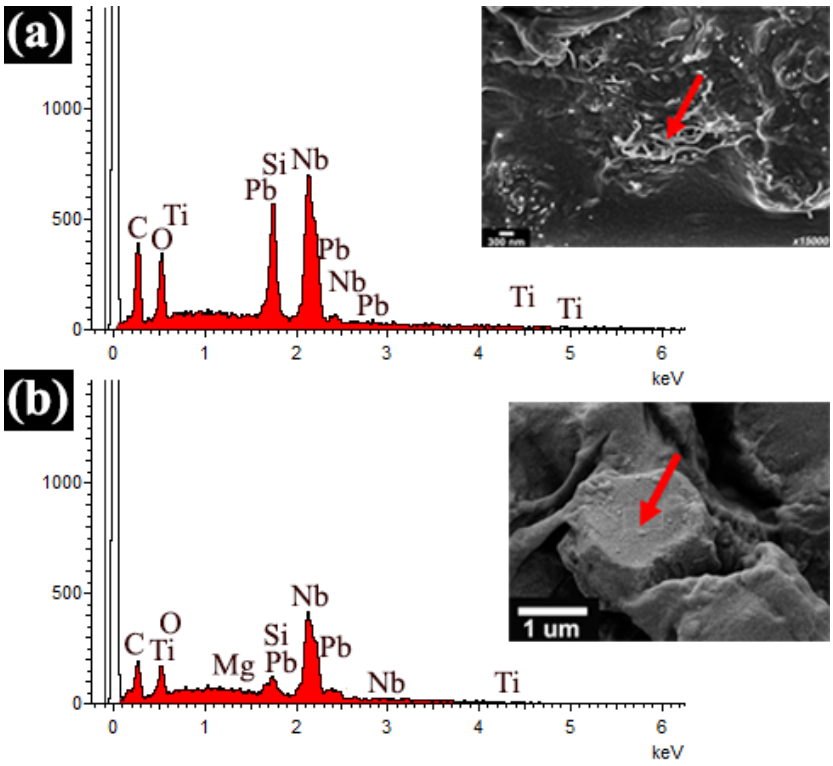

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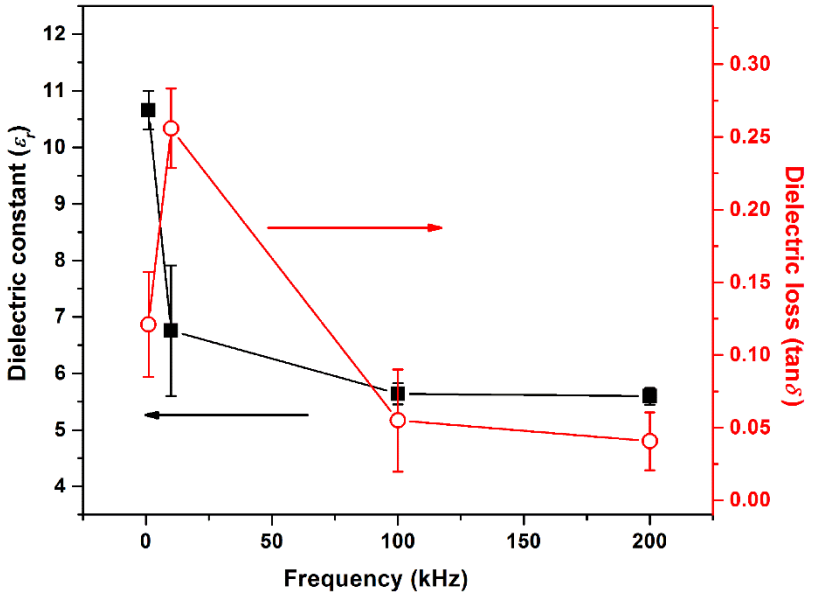
2 Fig. 5



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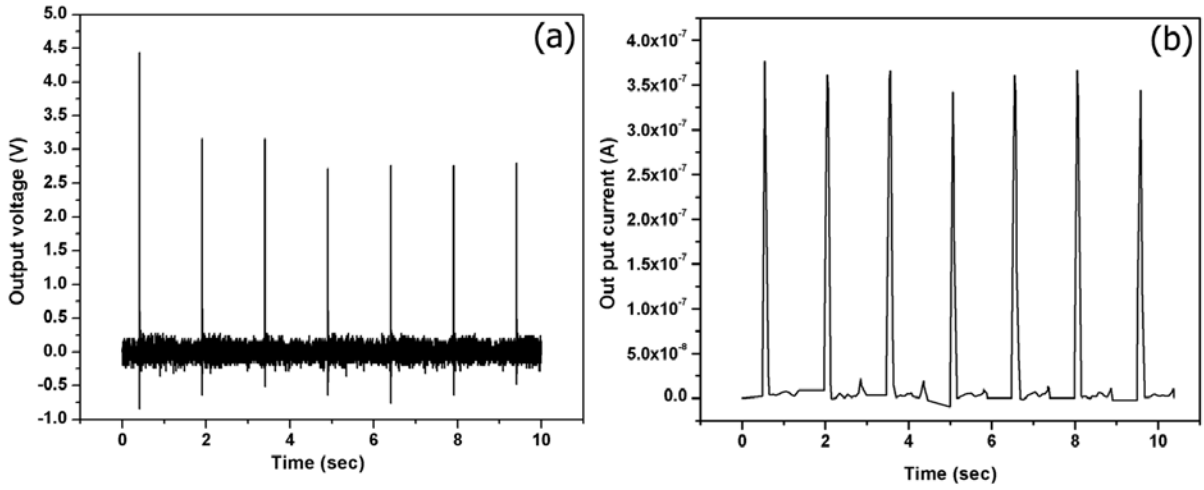
4 Fig. 6



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