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A high-sensitivity and fast-response nanocomposites-inspired sensor for acousto-ultrasonics-based structural health monitoring

Yaozhong Liao

Mechanical Engineering, Hong Kong Polytechnic University, Hung Hom, Kowloon, 999077, HONG KONG; 14103846g@connect.polyu.hk

Feng Duan

CAS Key Laboratory of Nanosystem and Hierarchical Fabrication, National Center for Nanoscience and Technology, Beijing, Beijing, CHINA; duanf@nanoctr.cn

Limin Zhou and Zhongqing Su

Mechanical Engineering, Hong Kong Polytechnic University, Hung Hom, Kowloon, 999077, HONG KONG; mmlmzhou@polyu.edu.hk; zhongqing.su@polyu.edu.hk

Promoted by an innovative sensing mechanism, a flexible carbon nanocomposite hybrid sensor made of graphene and polyvinylidene fluoride (PVDF) has been developed. In virtue of the tunneling effect in the conductive network formed in the nanocomposites, the sensor can be used to perceive acoustoultrasonic wave signals with ultralow magnitudes in a broad frequency range. To advance the insight into the sensing mechanism, both the scanning electron microscopy (SEM) and X-Ray diffraction (XRD) are employed to explore the dispersion of nanofillers and the crystal characteristic of the sensor, respectively. The sensing ability of the developed sensor is testified through the acquisition of strain signals from low frequency cyclic tensile loading to high frequency ultrasonic guided waves. Based on excellent mechanical and electrical properties of graphene, the sensor, fabricated with a solution film-forming method, can reach a high gauge factor of ~ 60 , responsive to ultrasonic signals up to 300 kHz. Being light weight and chemically stable, the developed sensor can be coated onto or embedded into engineering assets with minute weight penalty and favorable environmental adaptation. The simplified fabrication process can significantly reduce the sensing cost while maintaining high sensing efficiency, benefiting ultrasonic-wave-based structure health monitoring.



1. INTRODUCTION

Sensors are the most crucial component in a structural health monitoring (SHM) or nondestructive examination (NDE) approach. Traditional piezoelectric sensors tend to be brittle, and they, together with wiring and cabling, may introduce significant weight penalty to a host structure provided a dense sensing network configured and adopted in practical implementation of SHM and NDE. This may restrict traditional piezoelectric sensors from being extended to specific applications such as aerospace structures.

Benefiting from excellent mechanical and electrical properties of nanocomposites, carbon nanoparticle-enhanced smart sensors have been a promising candidate to overcome the limitations of traditional piezoelectric sensors. Successful use of nanocomposites-inspired sensors includes detection of gas leakage,¹ measurement of static or quasi-static strains,² acquisition of dynamic responses,³ and detection of structural damage.⁴ Driven by recent advances in nanotechnology, a new breed of nanocomposite sensor, made of carbon black (CB) and polyvinylidene fluoride (PVDF), has been developed in our previous work.⁵ Due to the agglomeration of CB particles, the response of this nanocomposite sensor to dynamic disturbance (e.g., ultrasonic waves) is yet limited to signals with relatively high intensity only. Motivated by this, the previously developed nanocomposite sensor is further improved in this study by introducing two-dimensional (2D) graphene to replace CB particles to form a conductive network using an enhanced processing approach, whereby to make the sensor responsive and sensitive to vibration or ultrasonic signals with weak magnitude yet high frequency.

2. SENSOR FABRICATION AND CHARACTERIZATION

A. MATERIAL PREPARATION AND SENSOR FABRICATION

The PVDF powder (Kynar k721, with a density of 1.74 g/cm³, and a melting point of ~158 °C, supplied by ARKEMA Corporation) was selected as the matrix of the nanocomposite, and solved in 1-Methyl-2-Pyrrolidinone (NMP) (with a boiling point of 203 °C, supplied by XILONG SCIENTIFIC Corporation) with a weight ratio of PVDF to NMP of 20:80, at 70°C until the solution became transparent. Then graphene aqueous solution (with a weight percentage of 2.7% and a conductivity of $1 \times 10^5 \text{ S/m}$, supplied by CARBON VALLEY Corporation) was added to PVDF solution. The mixture was stirred by a low speed mixer for 2-hour at a speed of 800 rpm. Afterwards, the mixture was poured onto a glass plate and cured at 150°C for another 2-hour. After cooling down at room temperature in air, a nanocomposite film was achieved by peeling off the mixture from the glass plate, and the film was tailored to individual flake-like membranes measuring 10 mm × 7 mm × 0.2 mm. Subsequently, each membrane was silver-pasted with two electrodes using a dual-component (polymer/silver powder) adhesive (D05001, Beijing Emerging Technology Co. Ltd) to configure a functionalized sensor, as shown in Figure 1.

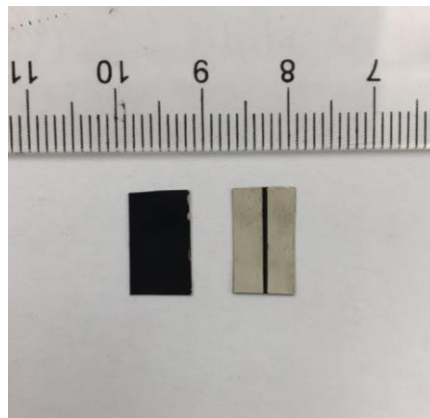


Figure 1. Fabricated sensors with silver-pasted electrodes (front and back sides)

B. ELECTRICAL PROPERTIES

The resistivity of the fabricated nanocomposite sensor changes dramatically when the content of nanoparticles reaches a certain weight percentage that is referred to as *percolation threshold*. When the weight percentage is close to the percolation threshold, most of the nanofillers are in a quasi-connected status, which means most of them are separated but extremely close to each other with a gap of several nanometers only. In this status, when the sensor receives an elastic disturbance (such as an ultrasonic wave signal), the variation in the nanoparticles distribution, even small, will induce a significant change in the resistivity manifested by the sensor, owing to the tunneling effect.⁶ The combination of piezoresistivity and tunneling effect endow the developed nanocomposite sensor with a capability to be responsive to elastic disturbance.

The resistivity (denoted by R , Ohm) of the fabricated nanocomposites was measured, at different degrees of graphene percentage in the nanocomposites (ranging from 0.3 wt% to 4.0 wt%), using a standard two-point volt-amperometric method with a digital multimeter (DMM 7510, Keithley). The electrical conductivity (denoted by σ , Siemens per meter, S/m) of the prepared nanocomposites was derived as

$$\sigma = \frac{L}{RA} \quad (1)$$

where L and A are the length and cross-sectional area of the fabricated sensor, respectively. Obviously, the conductivity changes dramatically when the graphene percentage is between 0.3 wt% and 1 wt%, on which basis the percolation threshold of the nanocomposite sensor was determined as 0.65 wt%, as seen in Figure 2.

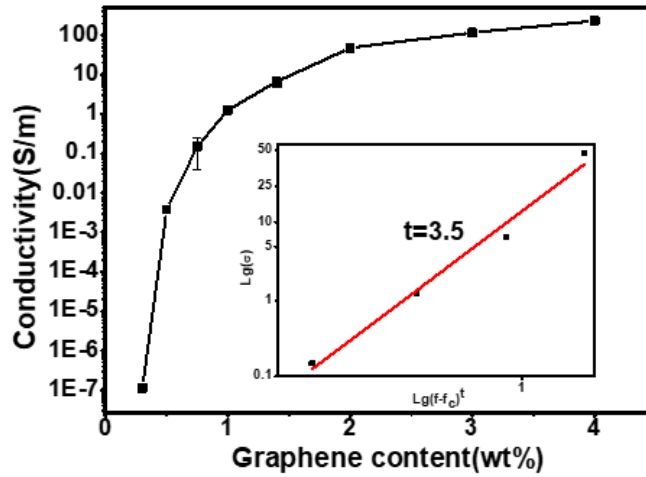


Figure 2. Conductivity of fabricated nanocomposite sensor at various degrees of graphene concentration

3. PROOF-OF-PRINCIPLE TESTS

A self-developed signal amplifier, with a built-in filter for increasing the signal-to-noise ratio, was configured to amplify the signals by converting change in resistivity to voltage. In some circumstances, the magnitudes of the signals, such as ultrasonic waves, could be ultra-low that are of the order of several micro-strains. The signals were captured with an oscilloscope (Agilent® DSO9064A).

A. LOW-FREQUENCY VIBRATION SIGNALS

A prepared nanocomposite sensor was glued on a glass fiber-epoxy composite cantilever (290 mm long, 38 mm wide and 2 mm thick), 70 mm away from the clamped end, to sense the low-frequency vibration signal induced by an electro-mechanical shaker (B&K® 4809, The Brüel & Kjær company) which was

driven by a waveform generator (7075, HIOKI company). The shaker excited the beam through a point 270 mm away from the clamped end with a sinusoidal vibration of different frequencies and magnitudes.

As representative results, Figure(a) shows the captured signals when the excitation frequency varied from 100 to 10000 Hz, to observe that the prepared sensor can be stably responsive to the applied vibration. A linear relationship between the magnitude of the response signals captured by the sensor and the magnitudes of the excitation can be observed, in Fig. 3(b), especially when the content of graphene is 1 wt% which approaches the percolation threshold.

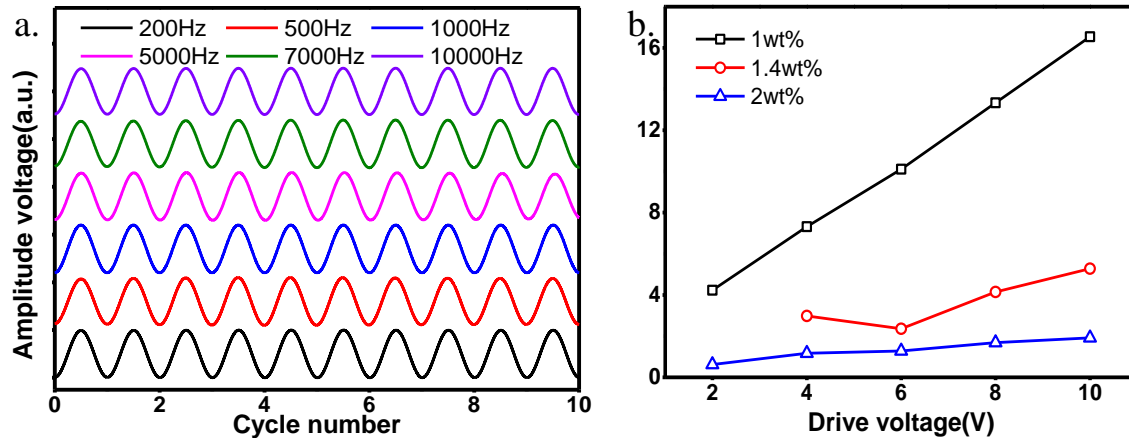


Figure 3. (a) Captured vibration signals under various frequencies (normalized by cycle number and amplitude); and (b) magnitude of captured signal versus intensity of excitation (using 200 Hz as an example)

B. HIGH-FREQUENCY ULTRASONIC GUIDED WAVE SIGNAL

Two piezoelectric wafers (denoted by PZT₁ and PZT₂, Physik Instrumente Co., Ltd., PIC151; diameter: 9 mm; thickness: 0.5 mm) were glued on a glass fiber-epoxy composite plate (600 mm×600 mm×2 mm) to emit and sense ultrasonic wave signals guided by the plate, respectively. Eight prepared nanocomposite sensors were surface-mounted onto the laminate panel, to form a sensor network for perceiving the signals.

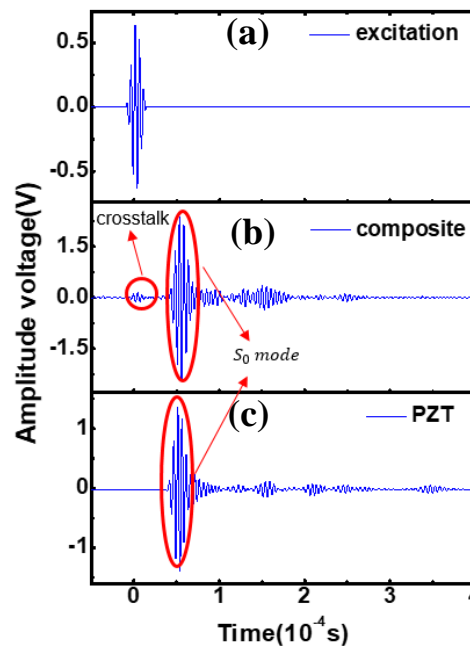


Figure 4. (a) Excitation signal (at 200 kHz); signals sensed by (b) graphene/PVDF sensor; and (c) PZT

Five-cycle Hanning-windowed sinusoidal tone bursts were excited to PZT₁ to emit ultrasonic guided waves at a central frequency ranging from 50 to 300 kHz with a waveform generator (PXIE-1071, National Instruments), after amplified to 400 Vp-p via the linear power amplifier (US-TXP-3, Ciprian Company). Figure shows the captured signals, when the waves were generated at 200 kHz, to observe that the zeroth-order symmetry Lamb wave mode (S_0) was acquired by the nanocomposite sensor without remarkable difference when compared with the signal sensed by PZT₂.

4. CONCLUSION

A high-sensitivity and fast-response nanocomposite sensor made of graphene and PVDF was fabricated. The sensing mechanism and sensing ability of the sensor were verified by broad-band frequency disturbance experiments (from low frequency vibration to acousto-ultrasonics signal up to 300 kHz). The signals are observed to be consistent with those acquired by traditional piezoelectric sensors without obvious discrepancies. Due to the 2D planar structure of graphene, this nanofiller has reduced possibility to aggregate compared with other candidate nanofillers such as carbon black and carbon nanotubes used in previous studies, which can be uniformly dispersed in the PVDF matrix to effectively induce tunnelling effect. This sensor can be networked to implement *in-situ* structural health monitoring.

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