

## Supporting Information

### **Active Control of Graphene-based Membrane-type Acoustic metamaterials by Low Voltage**

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#### **Materials and Methods**

##### **Materials:**

Poly(vinyl alcohol) was provided by Sigma-Aldrich. Graphene sheets were prepared by the liquid phase stripping method as previous work.

##### **Synthesis of PVA/graphene nanocomposite membrane:**

PVA/GR nanocomposite membranes were prepared by simple coat method. 15 g of PVA was dissolved in 300 mL deionized water at 95 °C for 2 h. Then, the different amount of graphene aqueous was added into the PVA solution under violent stirring to obtain a homogeneous mixture. Subsequently, the resulting mixture was casted onto a Cu plate equipped by plastic film and was coated into film using a rule. The thickness of film can be controlled by two same thickness Al strips with 20 cm\*4.0 cm placed on the both sides of Cu plate as a standard. The film was dried at 90 °C on a panel heater. Finally, the film was peeled off from plastic film substrate for further characterizations and applications. For comparison, neat PVA film, PVA/GR hybrid film with different amount of graphene

content consisting of 15%, 25% and 50% with respect to the PVA/GR composite were fabricated in the same way, respectively.

### **Characterization:**

Scanning electron microscope (SEM, model SU8000, Hitachi, Japan) was used to character the morphology and microstructure of nanocomposite membranes. X-ray diffraction (XRD) patterns of nanocomposite membranes were analyzed by Cu K $\alpha$  radiation (D/MAX-rA, Japan). Raman spectra were performed with a Lab RAM HR800 from JY Horiba. Fourier transform infrared (FT-IR) spectrums of composites was obtained using a model TENSOR-27 FTIR spectrometer from Bruker. The introduced fraction of graphene was investigated by Thermogravimetric analysis (TGA) using a Q5000 analyzer (manufacturer Q5000) from 30 to 800 °C under N<sub>2</sub> at a heating rate of 20 °C min<sup>-1</sup>. The electrical properties of the samples were measured by a four-probe method using Keithley 2400. The thermal conductivity of films was tested by TPS 2500S from Hot Disk at room temperature. Dynamic mechanical analysis (DMA) by temperature sweep of films were conducted at a constant frequency of 1 Hz, strain 1%, from 10 to 300 °C and a heating rate of 3 °C/min under tensile mode using a dynamic mechanical analyzer (DMA Q800, TA). Poisson's ratio for the films was obtained by measurement of transverse strain and axial strain combining using Force Transducer (DH3820) and Static Instrument (Instron 55298). Electromagnetic

interference (EMI) shielding effectiveness was obtained using a Vector Network Analyzer (Agilent Technologies N5227A, USA). The acoustic properties of nanocomposite membranes were obtained by using a modified impedance tube for the measurements of the transmission coefficients. The circular sample were fixed in the middle of two square impedance tubes. The both ends of the membrane were connected to a DC voltage, and acoustic data is recorded every 5V. From the transmission spectrum, the first anti-resonance frequency of nanocomposite membranes with different graphene content or under different voltage were observed and summarized. The vibration properties of the circle films with radius 1.5 cm were obtain by using a vibration shaker (YMC VT20).

**Table S1.** Weight fractions (theoretical and experimental) of the PVA/GR nanocomposite membranes

<b>sample</b>	<b>Theoretical weight fractions (%)</b>	<b>Experimental weight fractions (%)</b>
PVA	0	0
PVA/GR1 5	15	15.7
PVA/GR2 5	25	24.3
PVA/GR5 0	50	47.4
GR	100	86.3

