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## **Supporting Information**

## **Directional Freezing-Induced Self-Poled Piezoelectric Nylon 11 Aerogels as**

## **High-Performance Mechanical Energy Harvesters**

## Ashitha George<sup>†,‡</sup>, Harris Varghese<sup>†,‡</sup>, Achu Chandran<sup>\*,†,‡</sup>, Kuzhichalil Peethambharan Surendran<sup>\*,†,‡</sup> and E. Bhoje Gowd<sup>\*,†,‡</sup>

<sup>†</sup>Materials Science and Technology Division CSIR-National Institute for Interdisciplinary Science and Technology, Trivandrum 695 019, Kerala, India.

<sup>‡</sup>Academy of Scientific and Innovative Research (AcSIR), Ghaziabad 201 002, India.

\* Author for correspondence: E-mail: <u>bhojegowd@niist.res.in</u> <u>kpsurendran@niist.res.in</u> achuchandran@niist.res.in



Figure S1. SEM images of (a)isotropic nylon and (b) anisotropic nylon 11 aerogel.



**Figure S2.** Azimuthal intensity profiles of (200) reflection obtained from 2D WAXD pattern of anisotropic nylon 11 aerogel.



**Figure S3.** Deconvoluted WAXD patterns of (a)isotropic nylon 11 aerogel and (b)anisotropic nylon 11 aerogel.



**Figure S4.** (a) 1D WAXD pattern (b) and 2D WAXD pattern of 10 wt% anisotropic nylon 11 aerogel.

ID WAXD patterns of 10% anisotropic nylon 11 aerogels show peaks at 20° and 23.8° corresponding to the  $\alpha$  form and peaks at 6.2° and 21.6° corresponding to the  $\gamma$  form. 2D WAXD pattern shows isotropic rings which indicate that the orientation of polymer chains is not observed for nylon 11 aerogels with increased concentration. DSC thermograms show three melting peaks corresponding to the melting of  $\alpha$  and  $\gamma$  crystals. This further suggests that 5 wt% is the optimum nylon 11 concentration for preparing anisotropic aerogels.



**Figure S5.** DSC heating curves of isotropic and anisotropic aerogels in comparison with that of nylon 11 pellet.

The crystallinity was calculated using the equation.

$$X_c = \frac{\Delta H_m}{\Delta H_m^\circ} 100\%$$

 $\Delta H_m$  = Enthalpy of fusion x = Weight fraction, is the content of pristine polymer used in composites.  $\Delta H_m^\circ$  = Enthalpy of fusion for 100% crystalline polymer (206 J/g)

The crystallinity values obtained for isotropic and anisotropic nylon 11 aerogels from DSC were around 54 and 46%, respectively.



**Figure S6.** Temperature-dependent crystal transitions in anisotropic nylon 11 aerogel. The behavior of (a) 1D WAXD patterns and (b) *d*-spacings of anisotropic nylon 11 aerogels on heating from room temperature to 150  $^{\circ}$ C and on further cooling down to room temperature.



Figure S7. P- E hysteresis loop analysis of anisotropic nylon 11 aerogels along transverse direction.



Figure S8: (a)Permittivity and (b) tan  $\delta$  of isotropic and anisotropic nylon 11 aerogels in the frequency span of 1000 Hz to 10 MHz.



Figure S9. Image of the nylon 11 aerogel PENG.



Figure S10: Impedance measurements of isotropic and anisotropic nylon 11 aerogel based

PENGs.



**Figure S11.** Short-circuit current of (a) anisotropic and (b) isotropic nylon 11 aerogel-based PENGs.



**Figure S12.** Open circuit voltage of (a) isotropic and (b) anisotropic nylon 11 aerogel-based PENGs while reversing the connection.



**Figure S13.** Open circuit voltage of (a) and (b) short-circuit current of anisotropic nylon 11 aerogel-based PENG when force was applied along the transverse direction.



**Figure S14.** Open-circuit voltage of 10 wt% anisotropic nylon 11 aerogel-based PENG when force was applied along the freezing direction.



**Figure S15:** Open-circuit voltage generated from anisotropic PENG under various biomechanical forces such as single-finger tapping, multiple-finger tapping, and palm tapping.



**Figure S16:** open-circuit voltage generated by slow and fast tapping on the anisotropic PENG attached to the mouse.



**Figure S17:** open-circuit voltage of the self-powered vibrational sensor attached to the electric sewing machine.



**Figure S18.** SEM analysis of (a) vertical and (b) horizontal sections of anisotropic nylon 11 aerogel-based PENG after continuous 10,000 cycles of operation.



**Figure S19.** Open circuit voltage of anisotropic nylon 11 aerogel-based PENG after 6 months which shows the stability of the device.