Single-layer humidity-responsive membranes with advanced applications and moisture-electricity generation ability Ioanna Tzoumani¹, Konstantinos C. Andrikopoulos¹, Joannis Kallitsis^{1,2} ¹ Department of Chemistry, University of Patras, GR-26504 Rio-Patras, Greece; ² FORTH/ICE-HT, GR-26504 Rio-Patras, Greece Exploring sustainable energy has been an urgent challenge for human society due to energy shortage. Harvesting electricity from ubiquitous atmospheric moisture has emerged as a promising technology to address the energy shortage challenge. Tremendous amounts of gaseous water molecules are stored in ambient air, serving as an overlooked, huge water and energy source^[1]. *Moisture electricity generators (MEGs)* hold the potential to be an exciting next-generation platform for energy harvesting, because electricity can be generated directly through spontaneous moisture adsorption by functionalized nanomaterials^[2,3]. Centering on the goal of high performance and continuous electricity output, we introduce single-layer crosslinked membranes employing water-soluble functional polymers combined with various carbon nanostructures, which show high sensitivity to humidity^[4]. The fabrication process is cost-effective, scalable, and environmentally friendly. Properties of these hygroscopic materials, including their microstructure morphology, physicochemical, mechanical, and electrochemical characteristics, as well as their humiditydriven response, have been extensively studied. These self-standing composite membranes, capable of responding to humidity gradients, can likely enhance energy harvesting performance and unlock additional applications in the fields of actuators for non-contact human-machine interfaces and soft robotics. Preparation of the single-layered crosslinked membranes Moisture as the key factor for actuation performance 1. Sonication Structural characterization of the swelled membranes 2. Mix with PAA 3. Casting at RT 90/10 **Porous structure 30% CNTs** aq. solution 4. Crosslinking **✓ Moisture transport** channels at 120 °C SO_3 ✓ Water exchange with surroundings <u>20µm</u> P(SSNa-co-GMA40) **ATR-FTIR** characterization Time (min) ✓ Free-Standing OD: 2500 cm⁻¹ OH, COOH: 3330 cm⁻¹ ✓ Single-Layer Efficient and prompt absorption of water **✓** Composite 0,20 5% CNTs A(%) from its surroundings **CNTs-COOH** Membrane 0,15 0,10 ✓ Epoxy-carboxyl **Quick and reversible** reaction response to changes in 0,05 √ Hydrogen bonds environmental humidity 3500 2500 Wavenumber (cm⁻¹ **Electrochemical Properties** Responsiveness of crosslinked membranes to humidity In-plane conductivity via 4-point probe ر 0,5 الح I-V characteristics **Bending angles** 5% CNTs 2,0E-04 -**10% CNTs** (S/m) Mainly 100 120 µm 2 or 2 o'3 ,0E-04 -10-🔼 30 μm 120 µm ionic **30% CNTs** electronic **0**,0E+00 -0,1 CNTs % loading ↑ -1,0E-04 10% CNTs 5% CNTs 0,01 15 20 25 30 35 40 Time (s) 10 15 20 25 30 35 40 **Time (s)** -2,0E-04 **Current Flow** ↑ (CB) 0,5 0,001ن -0,5 0,0 0,5 10 15 20 25 30 **20% CNTs** Voltage (V) **30% CNTs CNTs** percentage **47 n.** 0,3 -**O**,3 Brightness of LED light increases with the CNTs content 90 µm 30 μm 90 μm _120 µm 20% CNTs 30% CNTs 20% CNTs 10% CNTs 5% CNTs $\alpha \times \pi$ $K = \frac{180^{\circ} \times L}{1}$ 10_15_20_25_30_35_40 Time (s) 5 10 15 20 25 30 35 40 Time (s) α (°): bending angle L (cm): length of the free section Lower thickness of the membranes of the curved membrane (|**3**,0x10⁻⁴ ¬ **Current flow for various RH%** Higher curvature and speed actuation 6,0x10⁻⁷ **Full flipping cycle** ′2,5x10⁻⁴ **Flipping** 20% CNTs --- 5% CNTs 10% CNTs 2,0x10⁻⁴ 4,0x10⁻⁷ ✓ On moist paper 10% CNTs 5% CNTs 1,5x10⁻⁴ ✓ On a bare hand 2,0x10⁻⁷ **♦** 0s ✓ With cargo ن 1,0x10⁻⁴ ✓ Different shapes -2,0x10⁻⁷ $5,0x10^{-5}$ ✓ Several thicknesses 80 100 **60** 40 50 60 70 80 90 Relative Humidity (%) **8s** 15s Film Thickness (µm) **Relative Humidity (%) APPLICATIONS** Moisture Carrying Soft robotics **MEGs Smart Gripper** cargo **Mechanism of moisture** electricity generator Ion diffusion Grasp Lift MEGs Transfer Na⁺ Release Swelled composite membrane Recover P(SSNa-co-GMA40) PAA / CNTs Contactless switches Membranes Vapors' source ~0,65 V **Biomimetic flower** 1x0,5 cm **Boiling water** Composite 0,6 -/oltage membrane 1x0,5 cm Humidifier 0,5 -Voltage 1x1,5 cm Water output TURNOR ~0,3 V vapors ~0,3 V Output 16 14 Multimeter Time (min) **Humidity on Humidity off** Humidifier of $\bigcirc 0,7 =$ Conclusions We developed humidity-sensitive CNTs-based membranes through a facile and environmentally friendly process. oltag Composite membranes: √ demonstrate rapid responsiveness to humidity changes. ✓ exhibit mixed conductivity, due to the incorporation of CNTs, and P(SSNa-co-GMA40) that exhibits ionic conductivity. 0,3 Asymmetrical electrode Ontput 0,2 0,0 0,0 ✓ with 30% CNTs can act as humidity-driven power-generator, providing a maximum voltage of ~600 mV. 0,2 configuration Acknowledgements References This research has been financed by the Greece 2.0-National Re-covery [1] Wang, P et al. Cell Rep. Phys. Sci. 2023, 4(8), 101517. and Resilience Fund: "Development of efficient third generation PV [2] Zhao, F. et al. Adv. Mater. 2015, 27(29), 4351-4357. 20 30 40 50 60 materials and devices to enhance the competitiveness of enterprises to [3] Xu, T. et al. Adv. Mater. **2024**, 36, 2209661. Time (min)

[4] Tzoumani, I. et al. J. Mater. Chem. C 2024, 12, 11594-11602.

the green energy produc-tion"_3GPV-4INDUSTRY. TAEDR-0537347.