Supplementary Information:

Real Time Chemical and Mechanical human motion monitoring with aerogel based wearable sensors

Onur Ergen\(^1,2,3\)*, Ecem Celik\(^1,2,3\), Ahmet Hamdi Unal\(^1\), Mert Yusuf Erdolu\(^1\), Feriha Eylem Sarac\(^1,2,3\), Ugur Unal\(^1,2,3\)

\(^1\)Department of Electrical and Electronics Engineering, Koc University, Sarıyer, Istanbul, 34450, Turkey.  
\(^2\)Graduate School of Science and Technology, Koc University, Sarıyer, Istanbul, 34450, Turkey.  
\(^3\)Surface Science and Technology Center, Koc University, Sarıyer, Istanbul, 34450, Turkey.

*Author to whom correspondence should be addressed: oergen@ku.edu.tr

1. Methods

1.1 GA/CNTs preparation. Graphene aerogels (GA) were prepared as in refs. 22 and 23 starting with graphene oxide (GO) precursors. 40mg GO was suspended in 3mL DI water and sonicated overnight. Then, ammonium hydroxide (NH4OH) was added to this solution, 1:5 by volume, followed by annealing at 80\(^0\)C for 24h to yield hydrogels. Any excessive solvent was then removed by supercritical drying to yield aerogel architectures, followed by a high temperature treatment at 1050\(^0\)C under argon for 2.5h. During the last 30min, 20sscm of nitrogen gas was
introduced to produce nitrogen doped graphene layers. The resultant aerogel structures were then broken down with small ball mills and loaded with functionalized carbon nanotubes with different mass proportions, as stated in the main text. These multi-wall carbon nanotubes (>98%, Sigma Aldrich) were functionalized by soaking them in a mixture of HNO₃:H₂SO₄, 1:3, at 90°C for 24h. Without this functionalization, CNTs only interact with Van der Waals force and the load transfer across the matrix is not effective. CNTs and GA are then dispersed in DI water for 6h and centrifuged at 3000rpm for 30min. Suspensions of this GA/CNTs mixture solution were filtered by vacuum filtration. The resulted membrane was then dried in air for 3h and vacuum filter dissolved in a NaOH solution followed by DI water soaking for 1h. The floating papers were then transferred on an Au coated PDMS film, followed by dying at 60°C for 3h. Finally, an acetone was dropped on the surface of the resultant aerogel structure at 30°C which greatly helps the triangular CNTs formation. This architecture is then infiltrated with liquid Sylgard PDMS@Silk (1% silk) at ambient temperature by leaving a contact region.

1.2 GA/CNTs microfluidic channel preparation. Microfluidic gaps were prepared during the filtering step, by blocking the filter paper’s pores horizontally, <1mm separation. These gaps were then filled with paraffin wax at 50°C, with a reverse filtration technique from the bottom so that wax can only fill the gaps, not the surface of GA/CNTs. After this step, regular PDMS@Silk infiltration occurred followed by wax removal with heat treatment under vacuum. The toluene solution was constantly dropped and vacuumed out during this step to remove all wax residues. The resultant paper was rinsed in DI water at 30°C and room temperature, repeatedly. Lastly, PANI electrochemically was deposited on exposed GA surfaces in these gaps as a monomer.

1.3 Characterization. The morphology of the GA/CNTs were examined using a scanning electron microscopy (ZEISS Ultra Plus Emission Scanning Electron Microscope). Due to the
fragile and soft nature of the composite material, only top view images were collected.

Mechanical tests were carried out using a simple tensile testing machine, with quasi-static and cyclic tensile cycles. The electrical conductivity was measured on the four-point probe and with a two-point method. The resistance variations and change of current were recorded by Keysight multimeter. Microfluidic measurements were carried out with Gamry potentiostat for validation, as a control experiment, and wireless data acquisition was carried out based on a Arduino, for a real-time readout. The sweat volume is measured in grams on a digital scale by change in weight and volume of the cotton. For real time measurements, we also used computer vision techniques to monitor cotton expansion and volume changing, then we correlated it with weight change. We prepared artificial sweat samples with different sodium ion concentration, with and without interfering ions, such as Cl⁻, K⁺, to properly train our ML algorithms (Fig. S7).
Figure S1 Ion concentration measurement of control experiments during exercise on three different subjects (forehead)
Figure S2 Measured sweat collection amount of control experiment #1 from subject’s forehead during exercise.
**Figure S3** Measured sweat collection amount of control experiments for machine learning training from subject’s forehead during exercise.
Figure S4 Resistance variation in typical a) torsion cycles, inset-electrical resistance vs cycle, b) bending cycles, inset- electrical resistance vs cycle, and c) compression test cycles.
Figure S5 a) Cycle performance of Reconstructed-GA/CNTs after the 1000th cycle. After the 1000th cycle, permanent thickness reduction was observed. b) Strain sensor performance with different angles.
**Figure S6** Simultaneous measurement of mechanical (bending of the elbow) and chemical sensing (Ion concentration).
Figure S7 Sodium ion calibration curves of artificial sweat samples. This sweat samples were prepared with Na\(^+\)-only and Na\(^+\) with interfering ions (Cl\(^-\) and K\(^+\)) at various concentrations. Multiple measurement, ~20, were taken at each Na\(^+\) concentration level in a interfering ion solution. A trend can be clearly seen that these measurements follows the Na\(^+\)-only solution control experiment.