Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information:

The water transport facilitated by carbon nanotubes enables a hygroresponsive actuator with a negative hydrotaxis

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Movie S1: The response of CS/MWCNTs film to moisture from the right.

Movie S2: The response of CS/MWCNTs film to dry N_2 from the right.

Movie S3: The response of CS/MWCNTs film on a moist substrate at 40 ℃.

Movie S4: The oscillating film.

Movie S5: A hanged strip of CS/MWCNTs functioning as a cantilever crane.

Movie S6: Composite film grabbing object droping object.

Movie S7: Motion of the device triggered by humidity stimulus.

Movie S8: A bi-responsive sensor which was assembled by a film, copper wire and two LED lights of different colors

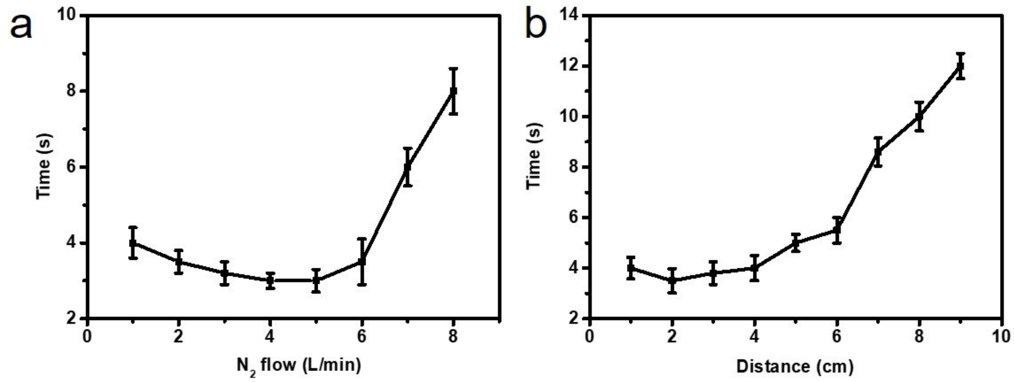
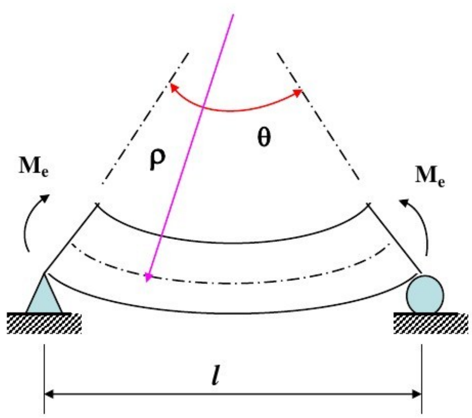


Fig. S1 Reversible transition properties of the actuator upon the gradient absorption/desorption of moisture at different nitrogen flow (a) and distance between the sample and the stimulus (b).

According to the equation below, the larger stain energy (u) during the bending deformation leads to larger kinetic energy after the removal of external force (e.g. the external response), and consequently induces faster recovery.



$$u = w = \frac{1}{2}M\theta = \frac{1}{2}\frac{\theta EI}{l}\theta = \frac{\theta^2 EI}{2l}$$

where, ρ is radium, θ is bending angle, l is the length of the sample, u is strain energy, w is work, I is moment of inertia of an area, M is bending moment, Me is external moment, E is modulus.

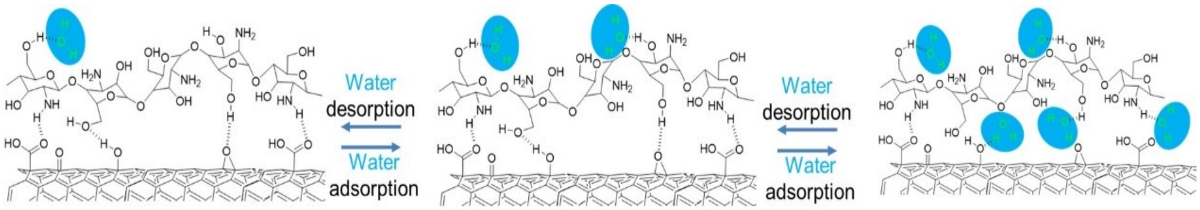


Fig. S2 Schematic structure of the composite film and mechanism of water exchange with environment. The structure changes (involving H bonds) in response to the adsorption and desorption of water (blue dots).

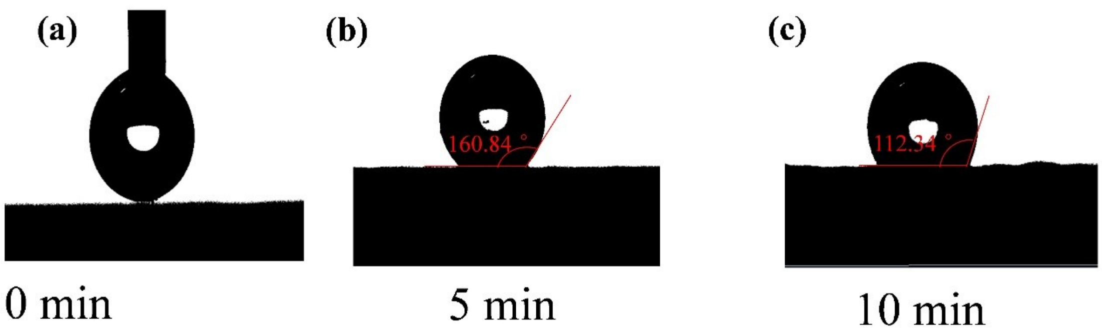
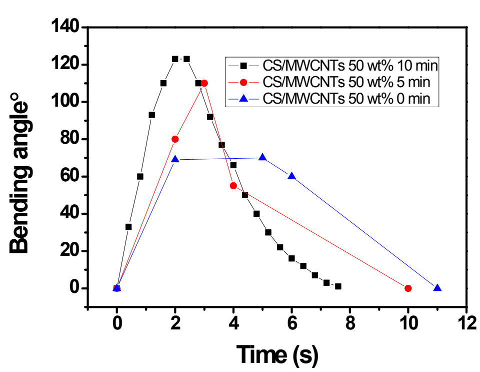
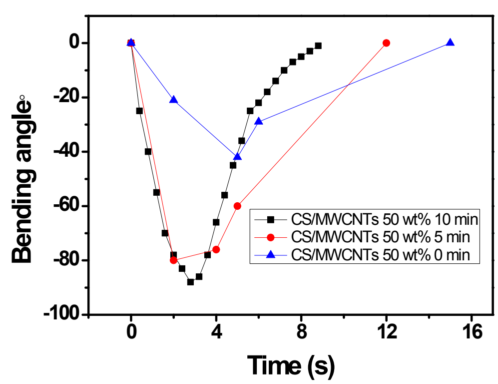


Fig. S3 Test of contact angles on MWCNTs upon varying durations of acid treatment.

The contact angle goniometer from Kruess was used to characterize the MWCNTs surface polarity. The contact angle measurements on the MWCNTs surface were carried out using a droplet of 5 μL at room temperature. As the final data, the mean of three values of contact angles on different areas of the membranes was used.



a



b

Fig. S4 Time-dependent angles of bending and unbending deformation of CS/MWCNTs 50 *wt*% composite film containing MWCNTs upon varying durations of acid treatment. (a) wet stimulus (RH=100%) and (b) dry stimulus (RH=0).

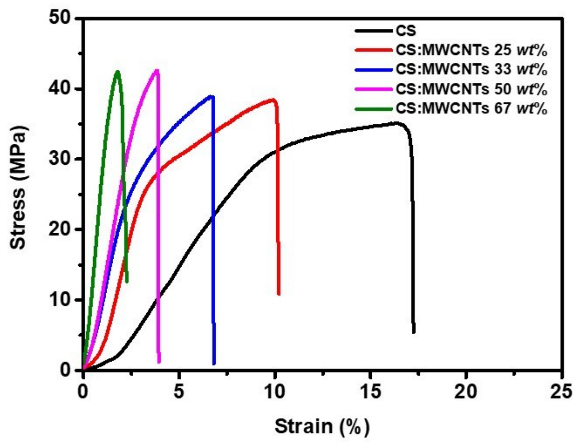


Fig. S5 Stress-strain curves of neat chitosan and its MWCNTs composite film with different MWCNTs content at a crosshead speed of 5 mm/min.

Tab. S1 Tensile modulus (E) and yield strength (σy) of chitosan/MWCNTs composites as a function of MWCNTs content.

Sample	Young's Modulus (E, GPa)	Tensile Strength (σ, MPa)	Elongation at Break (ε, %)
Pure CS	0.410	35.1	17.2
CS/MWCNTs (25 *wt*%)	1.05	38.4	10.2
CS/MWCNTs (33 *wt*%)	1.20	38.9	6.81
CS/MWCNTs (50 *wt*%)	1.57	42.6	3.93
CS/MWCNTs (67 *wt*%)	2.88	42.5	2.27

The typical stress-strain curves of chitosan and its MWCNTs composites at 50% RH are shown in Fig. 1. The summarized tensile properties are listed in Tab. S1. It can be seen that, the addition of MWCNTs significantly improves the tensile properties of chitosan matrix, and the mechanical properties increase with the increase of MWCNTs loading. With the 50% content of MWCNTs, the tensile modulus and strength of the composite increase dramatically by about 293% and 21%, respectively, compared with those of its neat counterpart. Due to the aggregation of MWCNTs within the matrix at higher concentrations, with further increase of the loading level of MWCNTs (67%), the tensile modulus increases slightly, while the tensile strength keeps stable. CS possesses three kinds of functional groups, amino groups, primary and secondary hydroxyl groups, in a glucosamine unit. The functionalized MWCNTs contain carboxylic and hydroxyl groups. Strong hydrogen bonds may be formed between CS and the MWCNTs.[43] The compatibility and strong interactions between MWCNTs fillers and the matrix greatly enhance the dispersion as well as the interfacial adhesion, thus significantly increasing the mechanical properties of the matrix.

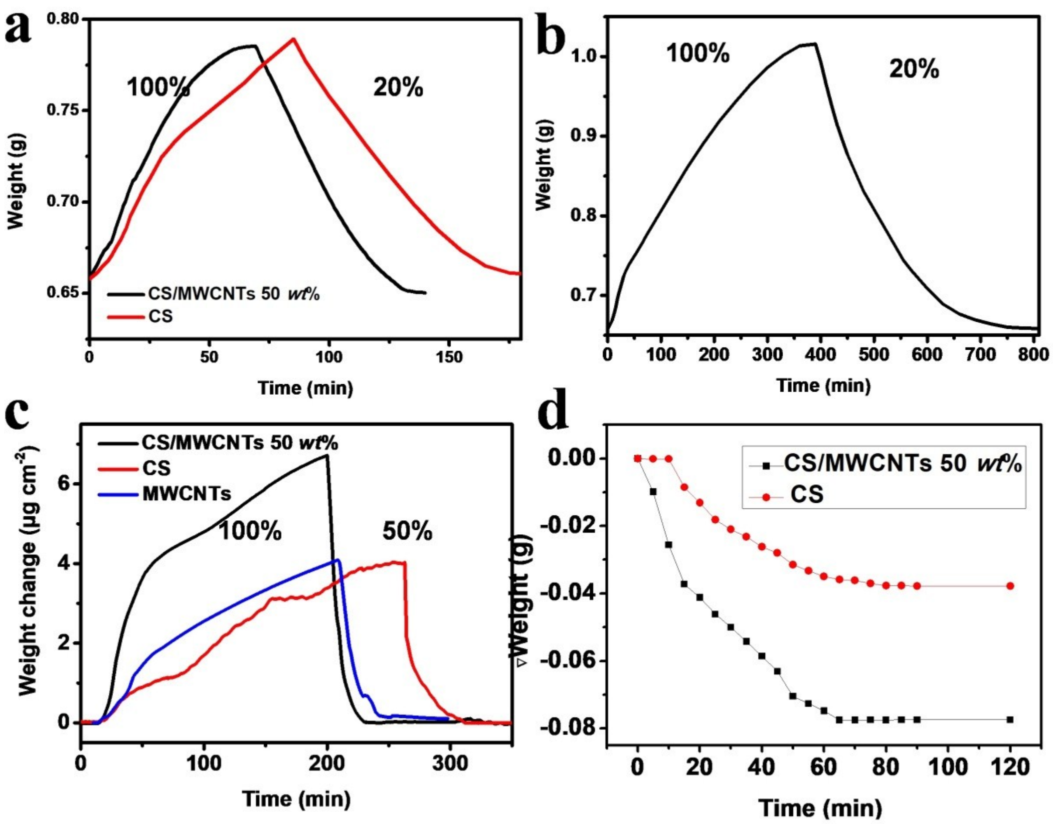


Fig. S6 (a) Time-dependent weight change of CS/MWCNTs 50 *wt*% (black) and CS (red) by weighing method upon increasing the humidity and then decreasing the humidity. (b) Time-dependent weight change of CS by weighing method upon increasing the humidity and then decreasing the humidity at longer time. (c) Weight change (monitored by QCM) as a function of time, upon the change of humidity from 50% to 100%: CS (red), CS/MWCNTs 50*wt*% (black), and MWCNTs (blue). (d) Time-dependent weight change of CS (red) and CS/MWCNTs 50*wt*% (black) by weighing method upon decreasing the relative humidity from 50% to 0.

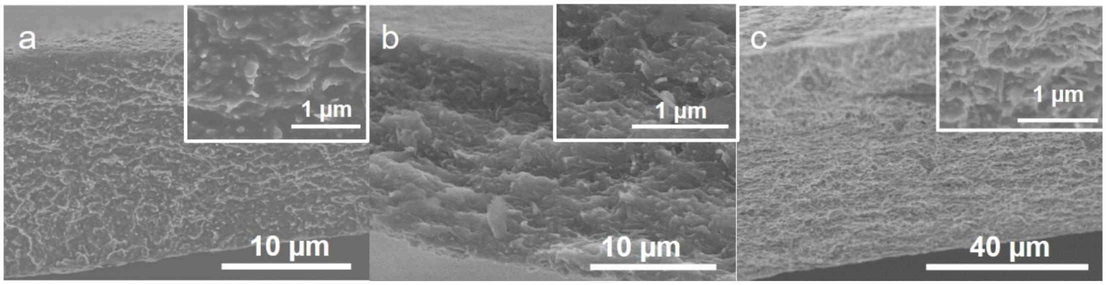


Fig. S7 Cross-section SEM image of the film with three weight contents of MWCNTs: (a) 0 *wt*%, (b) 33 *wt*%, and (c) 67 *wt*%.