## Ultrasensitive and Highly Compressive Piezoresistive Sensor Based on Biopolyol-Reinforced Polyurethane Sponge Coated with Silver Nanoparticles and Carbon Nanotubes/Cellulose Nanocrystals

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**Figure S1.** (a) Synthesis of bio-based polyester polyol. (b) <sup>1</sup>H NMR spectrum of biobased polyester polyol. (c) Synthesis of bio-based polyether polyol. (b) <sup>1</sup>H NMR spectrum of bio-based polyether polyol.

$$2 R - NCO + H_2O \xrightarrow{\text{DMDEE}} R^{-N} C_2^{-N} R + CO_2 \uparrow$$

**Figure S2.** The reaction equation for the reaction of isocyanate groups with moisture to form carbon dioxide.



Figure S3. Water contact angle of original PU sponge and TA-PU sponge.



Figure S4. Uniform black outer appearance and fractured surfaces of AgNPs/CNTs-CNCs@TA-PU sponge.



Figure S5. Green leaf stably supports the AgNPs/CNTs-CNCs@TA-PU sponge.



**Figure S6.** The relationship between the conductivity of the sponge and the dipping number.



Figure S7.  $\Delta R/R_0$  of the conductive AgNPs/CNTs@TA-PU sponge at different compressive strain.



Figure S8. TGA and DTG curves of AgNPs/CNTs-CNCs@TA-PU sponge.



Figure S9. Current-voltage characteristic curves of healed conductive sponges under different compressive strain.



Figure S10. SEM images of the healed AgNPs/CNTs-CNCs@TA-PU sponge.