Supplementary file

Preparation of NCDs particles: 4.2 grams of citric acid were dissolved in 40 ml of deionized water and subjected to ultrasonication for 10 minutes with stirring. After complete dissolution of citric acid, 1.34 ml of ethylenediamine was added and subjected to ultrasonication for another 10 minutes with stirring. Once the solute was fully dissolved, the mixed solution was transferred to a 200 ml stainless steel high-pressure autoclave lined with polytetrafluoroethylene (PTFE). The autoclave was heated at 200°C for 6 hours in an oven. After cooling the reaction vessel to room temperature, the solution was transferred to a centrifuge tube and centrifuged at 10,000 rpm for 5 minutes to remove impurities settled at the bottom. Subsequently, the brown liquid was subjected to dialysis in deionized water. During dialysis, the deionized water was replaced every two hours, and the dialysis process lasted for 48 hours. The dialyzed solution was then freeze-dried in a freeze dryer at a cold well temperature of -50°C for 72 hours to obtain purified NCDs particles.

Areal strain test: During area strain rate testing, the camera was directly fixed above the ring-shaped actuator, and the area and electric field of the electrode before and after power-on were photographed and recorded. Then, the image was processed

by ImageJ software. The calculation formula of area strain S_p is $S_p = \frac{(S_1 - S_0)}{S_0} \times 100\%$, where S_0 is the initial electrode area and S_1 is the electrode area after electric field driving.



Fig. S1 Preparation process diagram of NCDs@SiO₂/PDMS precursor solution.



Fig. S2 Contact angle test diagrams of (a) water, (b) NCDs and (c) NCDs@SiO₂ solution on PDMS film are respectively adopted, and the contact angle increases gradually in turn.



Fig. S3 Size distribution of NCDs particles.



Fig. S4 Photographs of NCDs@SiO₂/PDMS films with different concentrations under different lighting conditions. (a) natural light, (b) UV excitation under natural light, (c) UV excitation. Table S1. Summary and comparison of actuated performances of advanced DE composites.

Composites	Electric field (kV mm^{-1})	Maximum area strain (%)	Ref
10 wt% m-BT/SR4	12	26	1
TiO ₂ -PDMS (plasticized)	37	18	2
(MWCNT-Ecoflex)/Ecoflex	12.5	20.3	3
BAC2	70	118	4
PDMS186	7	4.2	This
NCDs@SiO ₂ /PDMS186	7	11.9	work



Fig. S5 Summary of area strain of DE actuators of different materials.



Fig. S6 Size and shape of the flexible PET frame.



Fig. S7 The dynamic bending Angle of a fluorescent crawling robot is shown in relation to (a) electric field and (b) frequency.



Fig. S8 Time series image of the motion of a fluorescent crawling robot.



Fig. S9 Fluorescent crawling robot's displacement over time curve.



Fig. S10 Optical image of a crawling robot on a field lawn.

References:

1 D. Yang, F. Ge, M. Tian, N. Ning, L. Zhang, C. Zhao, K. Ito, T. Nishi, H. Wang and Y. Luan, *J. Mater. Chem. A*, 2015, **3**, 9468–9479.

2 H. Zhao, D.-R. Wang, J.-W. Zha, J. Zhao and Z.-M. Dang, J. Mater. Chem. A, 2013, 1, 3140–3145.

3 Z. Xu, S. Zheng, X. Wu, Z. Liu, R. Bao, W. Yang and M. Yang, *Compos. Part Appl. Sci. Manuf.*, 2019, **125**, 105527.

4 L.-J. Yin, Y. Zhao, J. Zhu, M. Yang, H. Zhao, J.-Y. Pei, S.-L. Zhong and Z.-M. Dang, *Nat. Commun.*, 2021, **12**, 4517.