

Fast and Stable Redox Reaction of Dynamically Stretchable Pseudocapacitors Based on MnO₂/CNT Hybrid Electrodes

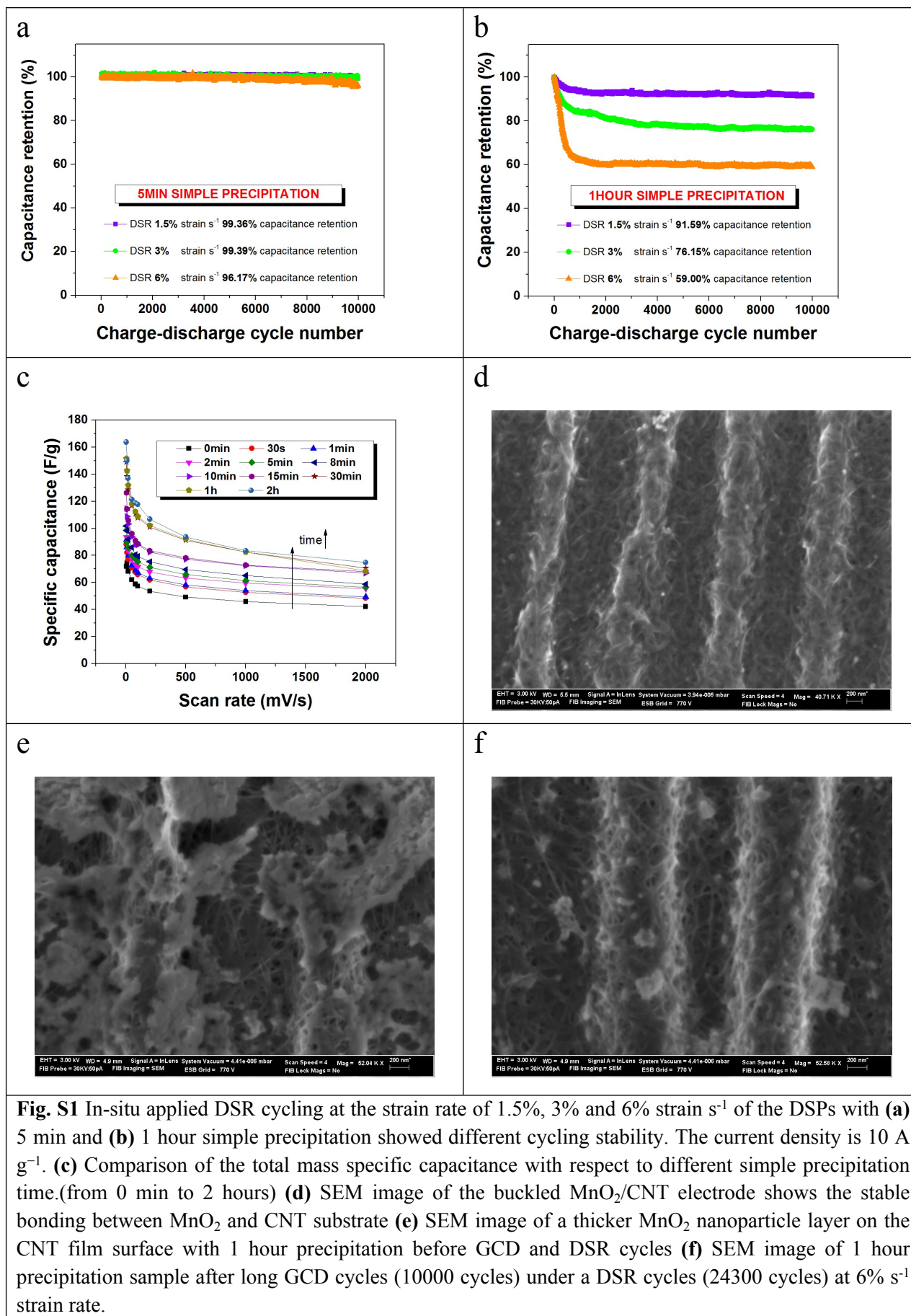
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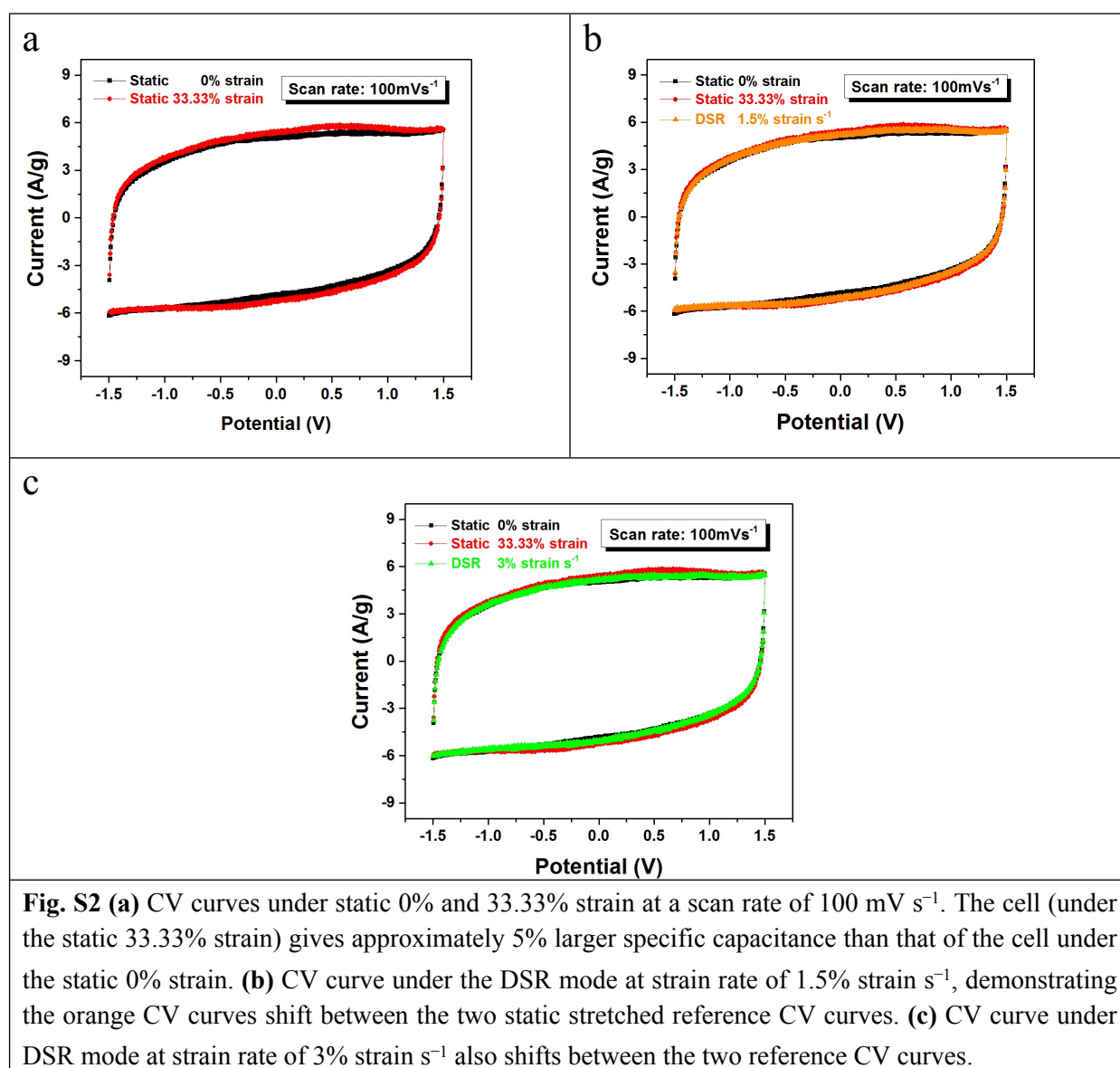
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Supplementary Materials

The MnO₂ mass loading on CNT films can be simply controlled by adjusting the precipitation time. The specific capacitance of the MnO₂/CNT based pseudocapacitors could be further increased by obtaining a thicker MnO₂ nanoparticle layer on the CNT film surface (Figure S1c, e). However, such a thicker MnO₂ nanoparticle layer will not hold a tight bonding between MnO₂ and CNT and leads to irreversible detachment of the MnO₂ nanoparticle layer when an intense mechanical force is applied (Figure S1b, f).



The specific capacitance of the MnO₂/CNT electrode is about 150 F g⁻¹ (based on the total mass of the MnO₂/CNT electrodes) and 750 F g⁻¹ (based on the mass of MnO₂ only) achieved at 2 mV s⁻¹, which then drops to about 120 F g⁻¹ and 600 F g⁻¹ respectively at 50 mV s⁻¹ (shown in Figure S1c). It is also interesting to note that even after 5 min precipitation, the specific capacitance of the hybrid film has been largely increased to 80 F g⁻¹, almost doubles the capacitance of the pure CNT based EDLCs (~45 F g⁻¹) at 50 mV s⁻¹, indicating that this fabrication process for the MnO₂/CNT hybrid films has the potential for being scaled up.



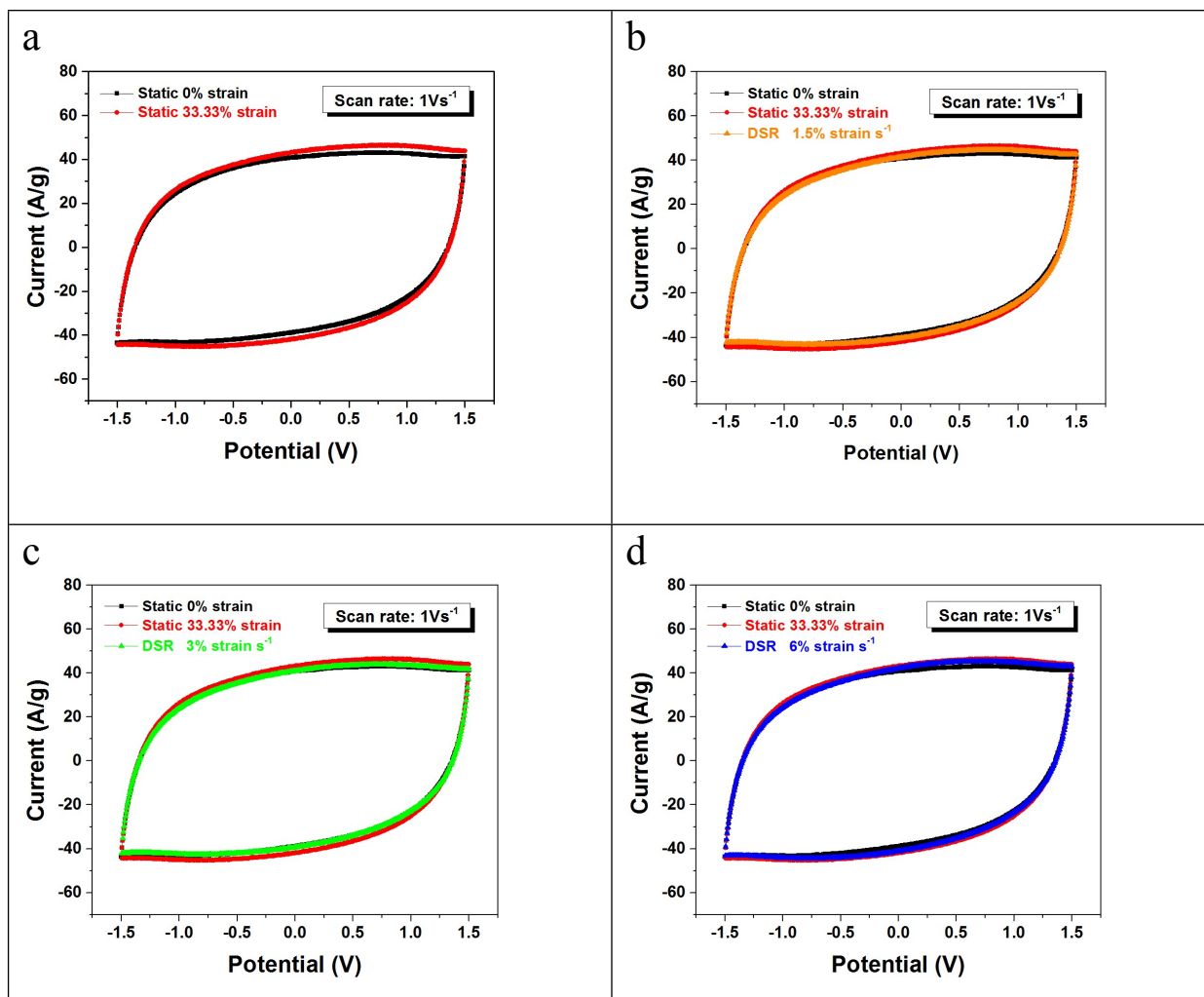


Fig. S3 (a) CV curves under static 0% and 33.33% strain at a high scan rate of 1000 mV s⁻¹. The cell still remains a rectangular shape, showing its good capacitive behavior, CV curve under the DSR mode at strain rate of (b) 1.5% strain s⁻¹, (c) 3% strain s⁻¹ and (d) 6% strain s⁻¹.

The specific capacitance is $\sim 91 \text{ F g}^{-1}$ at 10 A g^{-1} under the dynamic mechanical processes and the total capacitance is $\sim 21 \text{ mF}$, while the areal capacitance is between 4.65 mF cm^{-2} (4.5 cm^2 area without stretching) and 3.63 mF cm^{-2} (6 cm^2 area with 33.33% applied strain).

Energy and power densities are the two key determining factors for the practical applications of the stretchable pseudocapacitors. The energy density is $\sim 73 \text{ Wh kg}^{-1}$ at the power density of 70 W kg^{-1} (Figure S4b). It preserves 12 Wh kg^{-1} energy density as the power density increases to 61 kW kg^{-1} . The energy density of the stretchable pseudocapacitor is much higher than those of the flexible symmetrical CNT/PANI¹ (7.1 Wh kg^{-1}) and the

stretchable graphene/PANI electrode² (23.2 Wh kg⁻¹). To the best of our knowledge, this is the first report of dynamically stretchable pseudocapacitors with such excellent performance in terms of power and energy densities, as well as its long cycle stability.

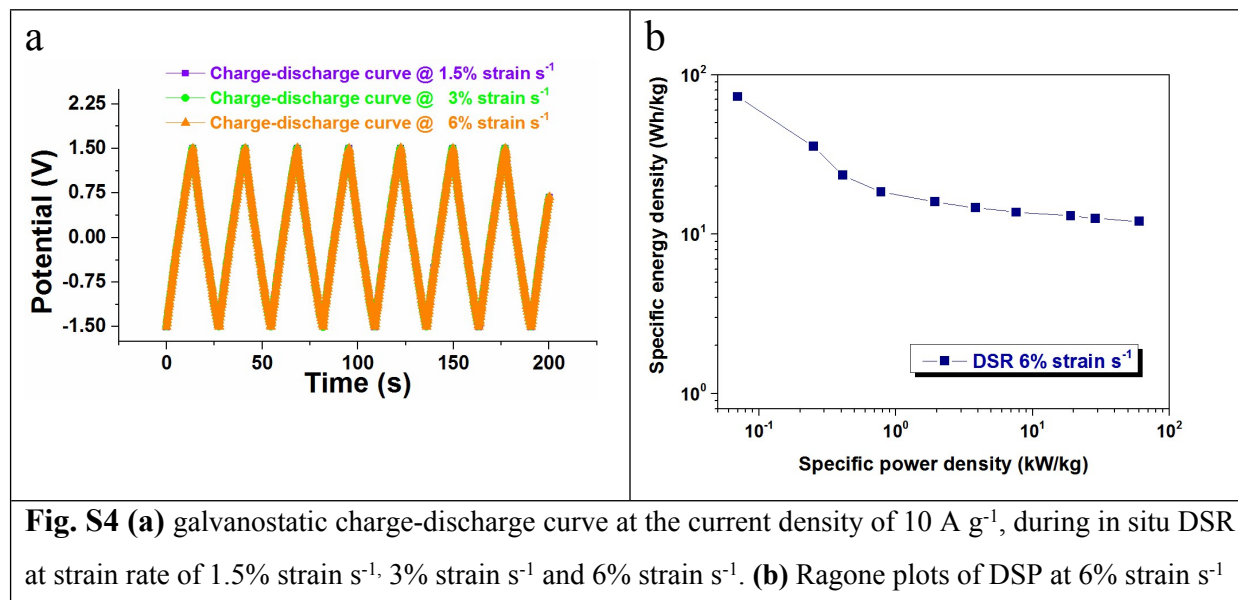
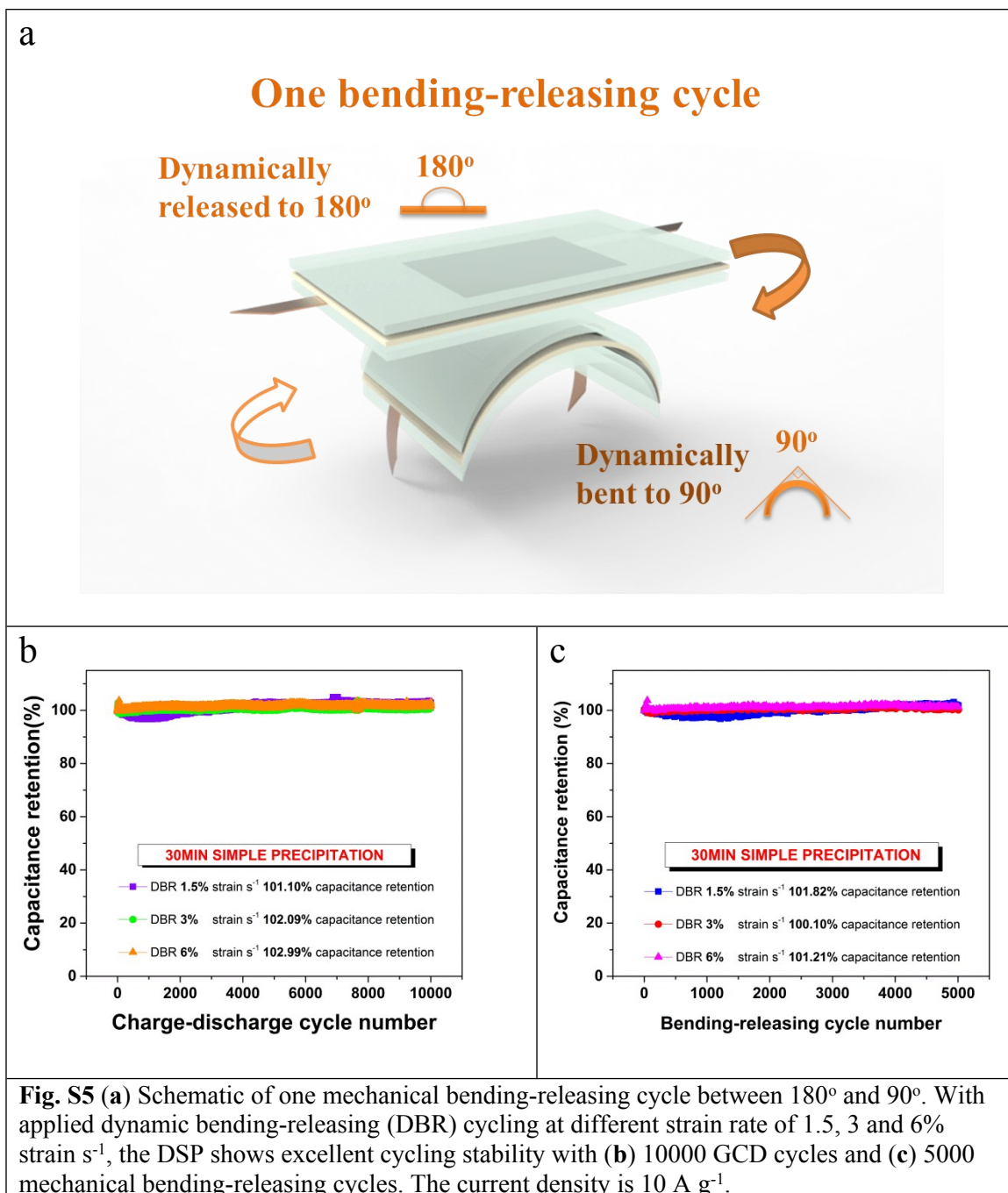


Fig. S4 (a) galvanostatic charge-discharge curve at the current density of 10 A g⁻¹, during in situ DSR at strain rate of 1.5% strain s⁻¹, 3% strain s⁻¹ and 6% strain s⁻¹. **(b)** Ragone plots of DSP at 6% strain s⁻¹

The bendability of DSPs is another determining factor when considering their huge potential market in applications such as roll-up display devices, smart sensors, e-papers, and flexible wearable electronics. Therefore, we conducted a flexibility test of our device at dynamically bending-releasing states that are different from static bending tests³. Here, we define one bending-releasing cycle as the cell is bent from 180° (flat) to 90° and then released back to 180° (as the schematic in Fig S5a). The capacitance retention of the DSPs under strain rates of 1.5, 3, and 6% strain s⁻¹ are shown in Figures S5b and S5c.



It is worth noting that different from the EDLCs/pseudocapacitors with a fading in capacitance under bending⁴, an excellent stability for DSPs under dynamic bending-releasing (DBR) was repeatedly observed, *i.e.* at different strain rates, about 1-2% improvement in the specific capacitance retention after 10000 electrochemical charge-discharge cycles and 5000 mechanical bending-releasing cycles is achieved, further proving the remarkable flexibility and stability of the DSP cell. This is likely the result of "mechanical-electrochemical synergy

activation", in which ions tend to adjust with cycling to fully access the active electrode material under the DBR process. Therefore, the DSP is not only stretchable but also bendable, demonstrating that DSP can potentially be applied in producing flexible electronic devices as a flexible power source.

Supplemental References

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