SUPPLEMENTARY INFORMATION

A Foldable Lithium-Sulfur Battery

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Supplementary Figure S1. Electrical conductivity of the CNT film, Single Side Electrode, Dual-Side Electrode, CNT/S-Single Electrode, CNT/S-Dual Electrode and 2CNT/S-Dual Electrode.

Method for force application and calculation of bending angle

Three point bending were simulated in our model. The force is labeled by black arrows in Supplementary Figure S2. In addition to the bending system in main text, we considered additional case where DWCNT being folded is under contact with single cross-linking DWCNT (Figure S2c). We want to use this model to test whether the number of cross-linking DWCNT could have an influence on the bending deformation. In the middle region, only the compression side was under pressure and the other tension side was free of external force. The forces were adjusted with the top and bottom regions so that the total force was zero and only torque was left. The bending angle θ is measured from the relative displacement d between top region and middle region, and the half length of the DWCNT length L by the equation

$$\theta = 180 - 2\cos^{-1}(d/L) \quad (1)$$

Supplementary Figure S2. Configuration of three DWCNT models studied herein. Three regions undergoing force application are labeled in green rectangle. The direction of force application are labeled with black arrow.

Plastic deformation for different DWCNT configurations

The onset bending angle at which plastic behavior began to set in for the first configuration (Figure S2a) was estimated to be 120° for the perfect hexagonal network of single CNT, which is consistent with previous reports (*Phys. Rev. Lett.* 76, 2511, 1996; *J. Chem. Phys.* 104, 2089, 1996). However, DWCNT bending showed a more complicated buckling pattern. Here we performed simulation of bending for single DWCNT with different maximum bending angle. When the DWCNT reached the maximum bending angle, it was relaxed for 20 ps under isothermal-isobaric (npt) condition. The final positions for the DWCNT after this relaxation process are gathered in Figure S3. We have used the same method in obtaining the final positions in Figure S4, Figure 1c-d. A kink or kink complex was formed at bending angle much lower than 120° , as seen in Figure S3. Plastic bending was assumed to have occurred when the CNT could not recover completely to its original position after 20 ps relaxation. From Figure S3, the onset of plastic deformation for DWCNT is found to be $\sim 150^\circ$. The onset of plastic bending was observed to be influenced by the bending force. In three point bending simulation, when the force was high enough, the onset angle for plastic deformation decreased to $\sim 130^\circ$ (Figure 1e). This is expected since application of higher bending force results in greater strain energy in the system at lower bending angle. In all cases studied here, this angle was found to be higher than 120° , which is consistent with the previous results of the elastic bending range of CNT (*J. Chem. Phys.* 104, 2089, 1996).

Supplementary Figure S3. Final position of DWCNT after full relaxation for different bending deformations. The degree below each subfigure is the maximum bending angle for this deformation. Corresponding with the formation of kinks, the residual bending angle (plastic deformation) begins to increase.

In order to further verify the influences of the number of cross-linking DWCNTs, single cross-linking DWCNT configuration was also analyzed for bending deformation. Figure S4 depicts the bending and recovery process for one DWCNT folded around another DWCNT. It is clear after undergoing close to 180° folding process, the DWCNT could not fully recover to its original position, and some plastic deformation could be seen. In main text we have discussed the condition in which one DWCNT is folded around two DWCNTs. In this last case studied here, the DWCNT was able to recover fully without any appreciable plastic deformation, as depicted in Figure 1c. This case can be considered representative of the experimental microstructure of the carbon nanotube film, as shown in Figure 1b. In reality, a combination of configurations shown in Figure S2b and Figure S2c may occur. These results show the fundamental mechanism responsible for elastic folding and unfolding of the CNT film observed in the experiments.

Figure S5 shows the map of strains in the CNT at the point of maximum folding for the configuration in Figure S2c. Due to the reduced number of cross-linking DWCNTs, the contact area was much smaller than for the configuration in Figure S2b. Therefore the strain concentration was still relatively high. Although no bond breaking was observed, the strain distribution was not as uniform compared to the results in Figure 1c of the main manuscript. Similar to three DWCNTs, kinks were formed at the back of this bending area. These kinks increase the area of the compression side, thus decreasing the compression strain at that region. However, for some atoms (atoms with red color in Figure S5) the strains were much higher than its neighbors. This abrupt change of strain caused the loss of original hexagonal network at some regions. As a results, after full unloading, such regions without periodic structure still exist, which causes the plastic deformation.

Supplementary Figure S4. Bending deformation of two DWCNT system. An obvious plastic deformation is confirmed after unloading. Different color is a representation of different atomistic strain and the color map is labeled on the right.

Supplementary Figure S5. (a) The back side of bending point for two DWCNTs (only the bended DWCNT is shown). The kinks formed during bending is labeled by black rectangle; (b) the front side of bending point for three DWCNTs. Different color is a representation of different atomistic strain and the color map is labeled on the bottom. From the comparison of different color at the bending region. The area with the loss of original hexagonal network structure is labeled by black rectangle. Different color is a representation of different atomistic strain and the color map is labeled at the bottom.

Supplementary Figure S6. EDS elemental mapping results of (a) sulphur, (b) carbon, (c) fluorine, and (d) oxygen.

Supplementary Figure S7. X-ray diffraction (XRD) patterns of sulfur-coated CNT film and pure CNT film.

Supplementary Figure S8. Raman spectra of the pure CNT film used as the current collector.

Supplementary Figure S9. Cyclic Voltammogram (CV) scans of the CNT/S-Single electrode at a scan rate of 0.1 mV s^{-1} for five cycles.

Supplementary Figure S10. Electrochemical performance of pure CNT film.

Supplementary Figure S11. SEM images of the lithium-metal surface after 200 cycles in the cell consisting of (a) CNT/S-Single, (f) CNT/S-Dual and (k) 2CNT/S-Dual electrodes. EDS mapping of the lithium anode surface after 200 cycles in the cell consisting of (b-e) CNT/S-Single, (g-j) CNT/S-Dual and (l-o) 2CNT/S-Dual electrodes.

Supplementary Figure S12. (a) The fabrication of the patterned sulphur electrode. (b) Photograph of the prototype foldable Li-S battery. The output voltage of the prototype foldable Li-S battery (c) under the flat (*i.e.* unfolded) condition and (d) after the application of two folds. The output voltage ($\sim 2.4\text{V}$) is unaffected by the folding.