Supporting Information

Carbon Welding by Ultrafast Joule Heating

Yonggang Yao,¹(a) Kun Fu,¹(a) Shuze Zhu,² Jiaqi Dai,¹ Yanbin Wang,¹ Glenn Pastel,¹ Yanan Chen,¹
Tian Li,¹ Chengwei Wang,¹ Teng Li,² Liangbing Hu¹,*

¹Department of Materials Science and Engineering, University of Maryland College Park, College
Park, Maryland, 20742

²Department of Mechanical Engineering, University of Maryland College Park, College Park,
Maryland, 20742

(a) Equally contributed

Email: binghu@umd.edu
**Pristine carbon nanofibers**

Figure S1 shows pictures and scanning electron microscopy (SEM) images of the carbon nanofibers (CNF) pre-carbonized at 873 K showing randomly intertwined fibers with a diameter of 200-300 nm.

![Figure S1](image-url)

**Figure S1.** (a)-(b) Images of pristine CNF in a flat and bent state. (c)-(e) SEM images of the pristine CNF at different magnifications.

We have deconvoluted the Raman peaks of pristine CNF as shown below (Fig. S2). It can be seen that the Raman peaks of region (800-1700 cm\(^{-1}\)) can be deconvoluted into 4 peaks: TPA (transpolyacetylene) band, D band, A band and G band. The deconvoluted G peak for pristine CNF is at 1590 cm\(^{-1}\), while the G peak for welded CNF is at 1585 cm\(^{-1}\). This red shift of G peak can be attributed to the increased graphitic degree in welded CNF. The A band is commonly observed in disordered carbon, and therefore is attributed to the point defects. For welded CNF, only D band and G band is present and clear, while the A band is suppressed. The deconvoluted Raman peaks again indicate the increased graphitic crystallinity in welded CNF as compared with pristine CNF.
Figure S2. Deconvoluted Raman peaks of pristine CNF.

**Joule heating process**

Figure S3 shows the high temperature light emitting effect induced by Joule heating at different powers. Figure S3a to S2d are images of the CNF film under Joule heating from 0 W to 1 W, 2 W and 3 W, with increasing brightness with increasing input power. Figure S3e to S2h show the light intensity change at 550 nm with 100 milliseconds exposure time at different temperatures.

Figure S3. (a)-(d) *In-situ* images and (e)-(f) thermal microscope images for the lighting process induced by Joule heating at different powers and temperatures.
Microstructures of pristine and Joule heated CNF

Figure S4 shows a high-resolution transmission electron microscopy (HRTEM) image of the pristine CNF with an amorphous carbon structure. No crystalline graphitic layer were observed in CNF matrix before Joule heating.

![HRTEM image of the pristine CNF film.](image)

**Figure S4.** HRTEM image of the pristine CNF film.

After Joule heating at 2800 K, a crystalline, graphitic carbon structure was observed in Fig. S5.

![HRTEM observation of the CNF after Joule heating.](image)

**Figure S5.** HRTEM observation of the CNF after Joule heating.
Figure S6 and S7 show the HRTEM images of the CNF film before and after Joule heating. The pristine CNF shows overlapping between two fibers at a junction point while for the Joule heated CNF film, clear graphitic carbon layers continuously connect two nanofibers.

Figure S6. HRTEM observation of pristine CNFs at a fiber junction. The white arrows indicate the fiber directions.

Figure S7. HRTEM observation of a Joule heated CNF at a fused fiber junction. The white arrows indicate the previous fiber directions. A continuous weld or fusion between two fibers is observed.
Figure S8 shows SEM images of the welded CNF at different magnifications.

![Figure S8](image)

**Figure S8.** (a)-(d) SEM images of the welded CNF.

**Composition analysis**

Figure S9 shows the composition change of CNFs film before and after the high temperature Joule heating. The N and O defects drastically decreased after Joule heating.

![Composition Analysis](image)
Figure S9. XPS analysis of the CNF film before and after the high temperature Joule heating.

The XRD of pristine CNF and welded CNF were measured and shown below (Fig. S10). The pristine CNF shows a broad peak in the range of 20-30 degree. In contrast, the welded CNF shows a very sharp peak at 26.5 degree, corresponding to the (002) peak. The sharp peak of welded CNF indicate increased crystallinity in welded CNF after high temperature Joule heating.

![XRD of pristine CNF and welded CNF](image)

Figure S10. X-ray diffraction pattern of pristine CNF and welded CNF.

CNT after Joule heating

Figure S11 shows the Raman spectra and conductivity measurements of the CNT film before and after the Joule heating technique. After Joule heating, the Raman D peak of the CNT film increased substantially, which means more defects are generated during the Joule heating process. This is in contrary to the Raman D band peak evolution of the CNF film after Joule heating. As a result, the conductivity of the CNT film decreased after Joule heating.
**Figure S11.** Raman spectra and conductivity measurements of the CNT film before and after Joule heating process.

**Conductivity measurement**

The CNF film electrical conductivity was measured at different powers as shown in Figure S12a. The resistivity was measured with a 10 mV bias voltage and then the current. Figure S12b and S12c shows the cross-section SEM images of pristine and welded CNF films used for the conductivity calculations. The films are thinner after Joule heating.

**Figure S12.** CNF film conductivity measurement results. (a) Voltage-current profiles of the CNF film after different Joule heating conditions (driving currents). (b) and (c) Cross-section SEM images of pristine and welded CNF films.
For single fiber conductivity measurements, the CNF film was first sonicated and then single fiber was transferred to the silicon wafer. A micrometer feature size shadow mask was used to measure the conductivity of this single fiber as shown in Figure S13a and S13b. The conductivity of a single fiber first went up and then decreased at excessively high current values. The maximum increase was around 180 times. The highest conductivity was calculated to be 822 S/cm.

Figure S13. Single fiber conductivity measurement. (a) and (b) Au electrodes on a single carbon nanofiber made by a micrometer sized shadow mask. (c) Single fiber voltage-current profiles under a bias field of 10 mV.

Polymer infiltration

Figure S14a shows strain-stress curves of pristine, welded, and polymer infiltrated CNFs films. Figure S14b and S14c show the cross sectional SEM images of the pristine and welded CNF films after polymer infiltration.
Figure S14. (a) The strain-stress curves for pristine CNFs, welded CNFs (w-CNFs) and welded CNFs after polymer infiltration (w-CNFs-epoxy). (b) and (c) Cross-sectional SEM images of the pristine and welded CNF after polymer infiltration.

Simulation details

We are effectively simulating the region that is at the intersection of two fibers, as shown in Figure S15(a). We first construct an initial structure, in which 6 parallel PAN chains on top (representative of the surface area of Fiber 1) are in perpendicular to another 6 parallel PAN chains on the bottom (representative of the surface area of Fiber 2), as shown in Figure S15(b). We used the large-scale atomic/molecular massively parallel simulator (LAMMPS) and REAXFF potential to perform molecular dynamics simulations. The time step is set to 0.25 fs. The simulation is performed on a canonical ensemble (NVT), controlled by a Nosé-Hoover thermostat. We first put the structure of Figure S15(b) into a relatively larger simulation box. The in-plane two directions are subjected to periodic boundary condition while the one out-of-plane direction is subjected to a reflected wall, as a way to introduce the excessive vibration of the atoms. We then equilibrate this structure at 300 K while gradually shrinking the simulation box size to 3.8 nm by 3.8nm by 0.68 nm within 50000 time steps. We make the out-of-plane simulation box size small so that the effect from the reflect wall at
the box boundary could be maximized on the entire structure, when considering that the electrons are passing through the simulation box from out-of-plane direction. The equilibrated structure is shown in Figure S15(c) and (d). The system has a density of 2.1 g/cm³, which is relevant to the experimental carbonization conditions. After the equilibrated structure is obtained, we use this structure for the input coordinates for subsequent simulations. Due to the limitation of time scale at any MD simulation, we are not able to match our simulation to the experimental heating rate. Instead, this structure is directly subject to a canonical ensemble (NVT) at 3000 K, controlled by a Nosé-Hoover thermostat, for 800000 time steps. Then we have obtained the structure as shown in Figure 4(b) in main text. After that, we manually delete the isolated nitrogen molecules/atoms and rerun the simulation. We repeated this process until all nitrogen molecules/atoms are removed. Then we use the same protocol on the subsequent removing the isolated hydrogen molecules/atoms.

**Figure S15.** Simulation strategy. (a) Schematic of simulated region: the contact area of two adjacent fibers. (b) The initial structure of CNFs and (c) top view and (d) side view of equilibrium CNFs structure at 3000 K.
Battery current collector demonstration

Figure S16. Demonstration of welded-CNFe membrane applied as a battery current collector. (a) Photo images of cathode slurry casted on w-CNFe film 0 and 2 minutes. (b) Sheet resistance of w-CNFe, w-CNFe loaded with LFP and a commercial LFP cathode material. (c)-(d) SEM images of LFP cathode infiltrated into w-CNFe as a battery current collector. (e)-(f) SEM images of higher loading of LFP on w-CNFe membrane.

As shown in Fig. S16a, the as-casted cathode slurry sits on w-CNFe membrane with a good wetting angle of ~ 30°, and quickly infiltrate into w-CNFe membrane within 2 minutes with the assistant of vacuum, indicating that the porous structure of w-CNFe membrane is suitable as a 3D current collector to incorporate micro- and nano-powders like LFP. Due to the unique
3D interconnected carbon structure with a continuous electron conductive pathway, the sheet resistance of w-CNF loaded with LFP cathode materials is still as low as 2.35 Ω/sq (Fig. R6b). In comparison, the commercial LFP cathode sheet (MTI) has a sheet resistance of 310 Ω/sq measured from in-plane LFP side, addressing the advantages of 3D current collector. The microstructures of the w-CNF-LFP composite are shown in Fig. S16c-d. The cross-section view shows the infiltration of LFP cathode material into w-CNF film. This is clearly displayed in Fig. S16d, that LFP powders are embedded into the 3D conductive w-CNFs scaffold. Such a w-CNF-LFP composite greatly reduces the conductive distance and therefore the resistance during electrochemical reactions. For higher mass loading, the SEM images are shown in Fig. S16e-f. From the cross-section view, the LFP electrode has a thickness of 45 µm while the w-CNF film is 15 µm, resulting in a mass loading of 4.8 mg/cm².