Electronic Supplementary Information

Photo-thermally controlled Cu nanoparticles density in SWCNT/Cu nanocomposites-based flexible EMI shielding electrodes

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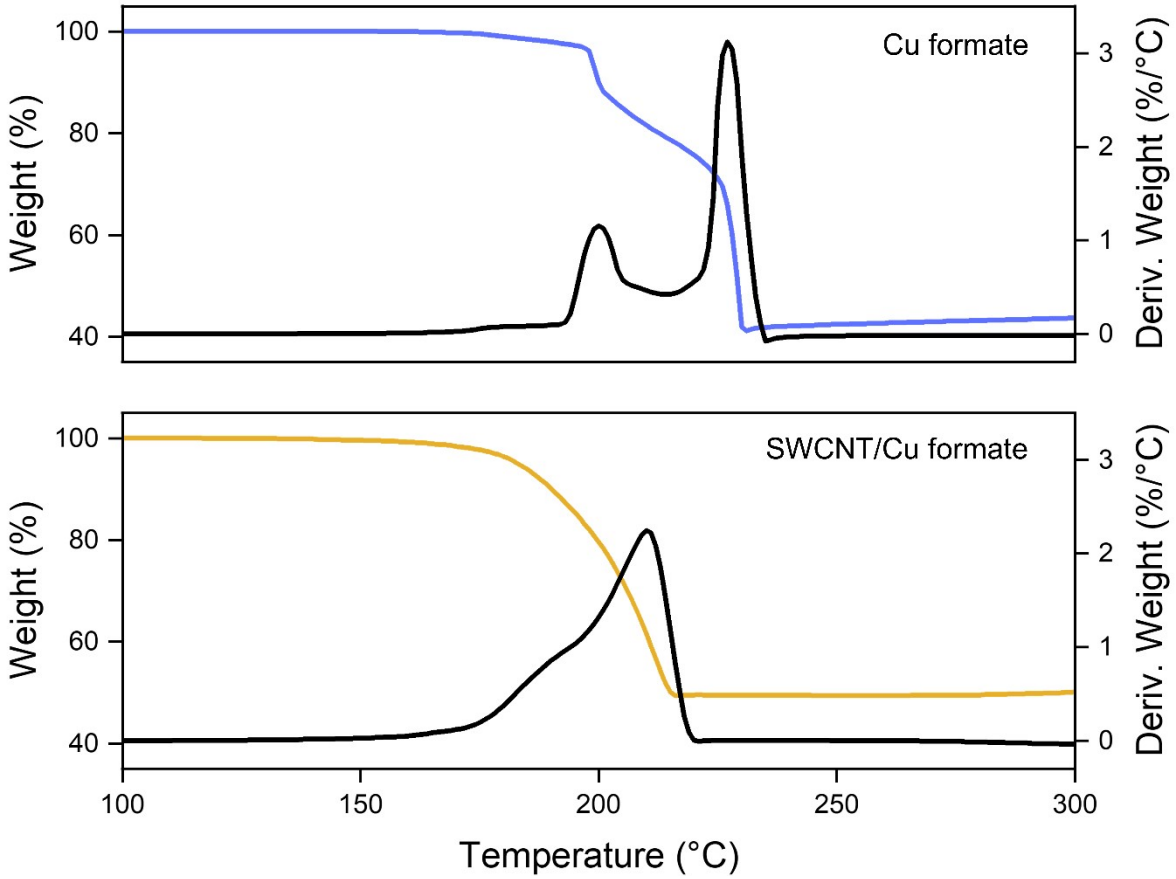

Fig S1. (a) DSC curves of (upper panel) Cu formate and (lower panel) SWCNT/Cu formate, obtained through TGA analysis.

We conducted TGA-DTG analysis to determine how the thermal decomposition behavior of the SWCNT/Cu formate composite compared with that of Cu formate alone. As shown in Fig. S1, the TGA-DTG curves showed a two-step weight loss in air, which was in accord with the decomposition mechanism of Cu formate proposed by Farraj et al. (Eqn. (1,2)).

(1) $Cu(HCOO)_2 \rightarrow Cu(HCOO) + CO_{2(g)} + {}^{1}_{2}H_{2(g)}$

(2) $Cu(HCOO) \rightarrow Cu_{(s)} + CO_{2(g)} + {}^{1}_{2}H_{2(g)}$

Copper formate decomposes through a two-step reduction process: the first step involves a weight loss up to 198 °C, which is attributed to an exothermic reaction, and the second step is terminated near 226 °C, indicating that copper ions are fully reduced to metallic copper through

an endothermic reaction. Notably, the composite showed a significantly lower weight loss temperature, which ranged from 180 °C to 210 °C and which indicated that decomposition occurred at lower temperatures. The relatively low decomposition temperature of Ox-SWCNT/Cu formate is attributed to nucleation occurring more readily in this composite compared with that in Cu formate, which had a high nucleation energy barrier. A homogeneous nucleation process occurred in the composite, in which nuclei were uniformly formed throughout the initial state since barriers to nucleation were overcome. The barriers could be significantly reduced since the Ox-SWCNTs acted as substrates. Additionally, the final mass increased by approximately 6%, which could be attributed to the incorporation of SWCNTs. These results indicate that the added SWCNTs acted as a complexing agent, promoting the reduction of Cu formate and thereby lowering the nucleation temperature, which enhanced the thermal stability of the composite.

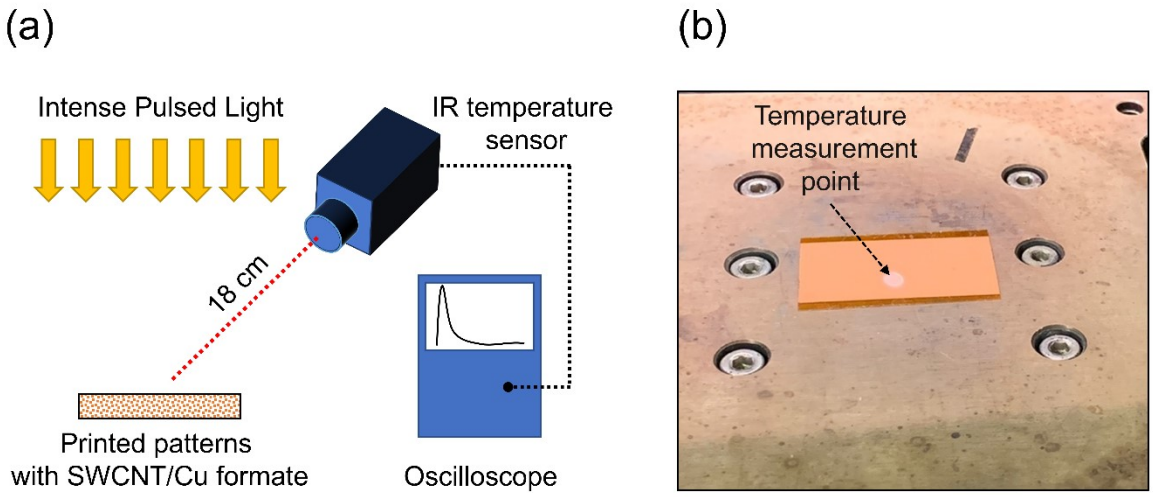

Fig S2. (a) In situ temperature monitoring system used during IPL irradiation. (b) A photograph of a surface temperature measuring point on a sample subjected to IPL irradiation.

During the *In-situ* temperature measurements, we monitored the temperature profile by using a non-contact ultrafast temperature sensor. The distance between the IR sensor and the sample was set to 18 cm, and the signal was monitored using an oscilloscope. This sensor helped to accurately capture temperature changes occurring during the IPL irradiation, which were important for understanding thermal effects on the loading of copper onto the surface of SWCNTs. Through this approach, we monitored the temperature profile accurately in real-time during the reduction of the copper ink, and we used the temperature profile to analyze the effect of temperature changes on the overall process effectively.

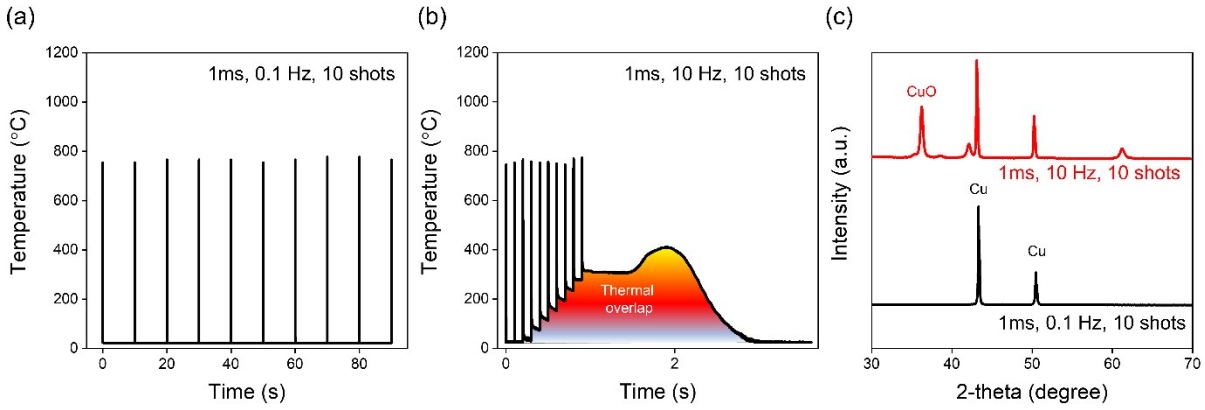

Fig S3. Measured temperature variation of printed Cu patterns during multipulse irradiation with IPL for pulse repetition rates of (a) 0.1 Hz and (b) 1.0 Hz. (c) X-ray diffraction patterns of Cu patterns sintered using IPL with pulse repetition rates of 0.1 and 10 Hz.

At higher repetition rates, we observed that thermal overlap indeed occurred, leading to an increase in the cumulative temperature. When thermal overlap caused the temperature to rise above 200 °C, the newly formed Cu nanoparticles (Cu NPs) could be oxidized. In other words, the high temperature was conducive to the oxidation of Cu NPs, which could lead to changes in or the degradation of the NPs and thereby affect their structural integrity and performance. Hence, the repetition rate should be controlled to prevent such adverse effects.

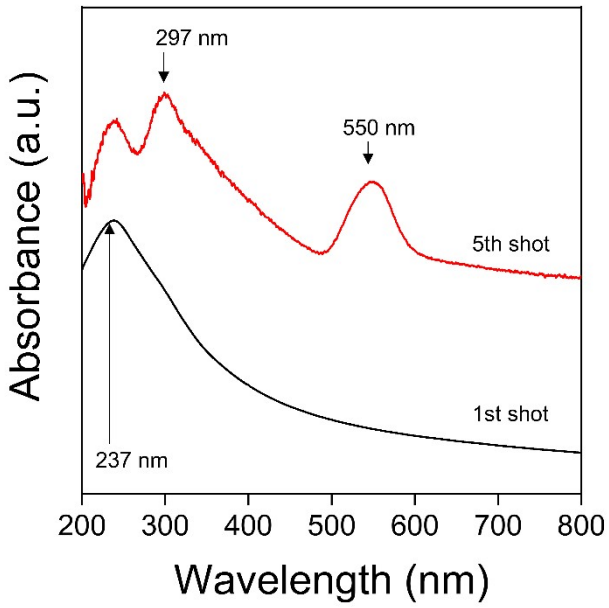

Fig S4. The UV-Vis absorption spectra of 1st-shot SWCNT/Cu and 5th-shot SWCNT/Cu composite.

The UV-Vis absorption spectrum of each sample shows a strong absorption peak in the UV region, which originates from the π-π^* transition of the aromatic sp^2 domain. The absorption peak undergoes a redshift from 232 to 297 nm upon the photothermal reduction of the Ox-SWCNTs. This shift is due to the restoration of sp^2 conjugated structure and increasing electron concentration.[1] The broad shoulder at around 550 nm corresponds to the newly formed Cu NPs.

1. M. K. Rabchinskii, A. T. Dideikin, D. A. Kirilenko, M. V. Baidakova, V. V. Shnitov, F. Roth, S. v. Konyakhin, N. A. Besedina, S. I. Pavlov, R. A. Kuricyn, N. M. Lebedeva, P. N. Brunkov, A. Y. Vul', *Sci. Rep*. 2018, **8**, 14154

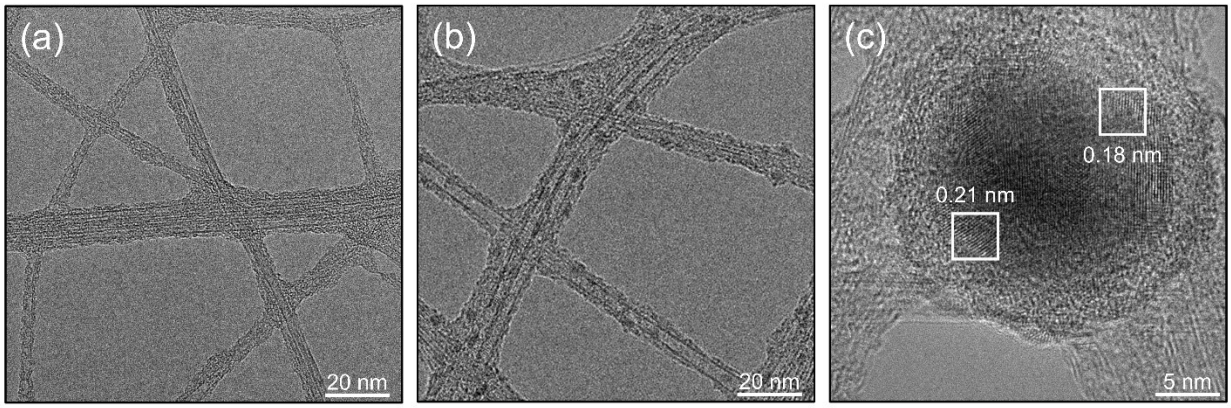

Fig S5. TEM image of (a) Ox-SWCNTs and (b,c) 4th-shot SWCNT/Cu composite.

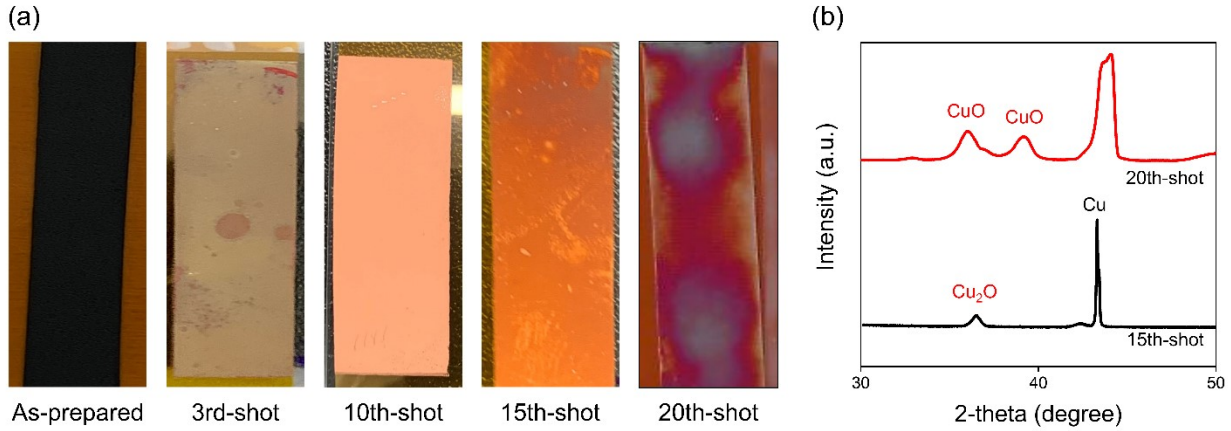

Fig S6. (a) Photographs of SWCNT/Cu composite patterns prepared with different numbers of IPL shots. (b) XRD results corresponding to the 15th-shot for SWCNT/Cu composites.

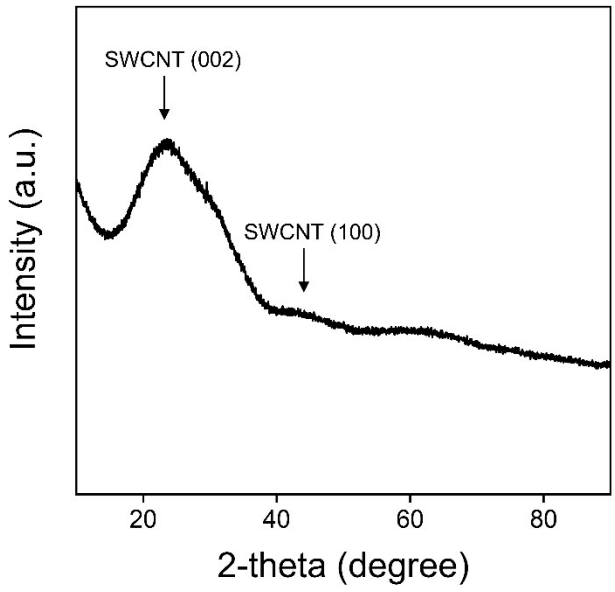

Fig S7. XRD data of Ox-SWCNTs

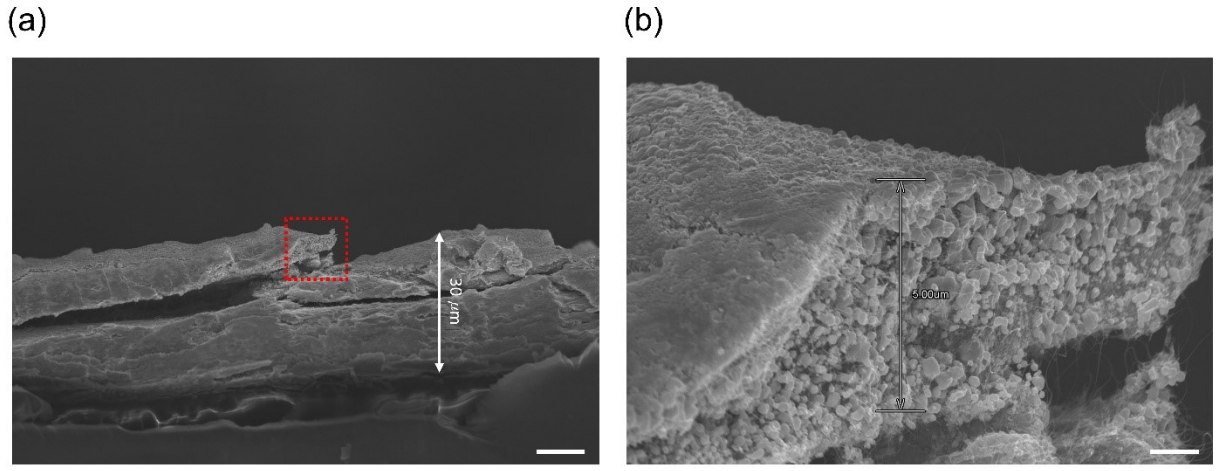

Fig S8. (a, b) Cross-sectional FE-SEM images of 10-shot SWCNT/Cu films (1:20 wt%). Scale bars are 10 μm (left) and 1 μm (right).