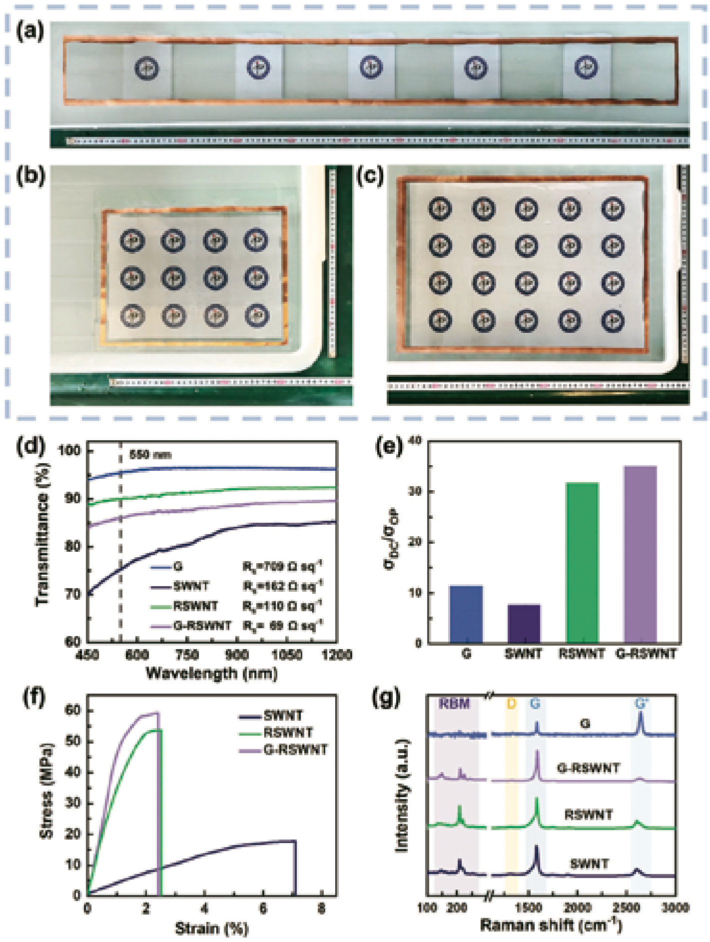
# Large-Area Preparation of Flexible Carbon Nanofilms with Synergistically Enhanced Transmittance and Conductivity

Large-area flexible transparent conductive films (TCFs) are urgently in great demand for future electronics, optoelectronics, energy devices, and applications in other fields. Indium tin oxide (ITO) TCF, which is widely used in modern technology, is difficult to meet the needs of scientific and technological development (especially the need for a new generation of flexible electronic devices), because indium is a non-renewable resource and expensive, and ITO is inherently brittle.

Currently, carbon nanofilms, metal nanowires, conductive polymers and other transparent conductive materials have been developed to replace ITO. Among them, the carbon nanofilm is considered to be one of the most promising candidates due to its excellent electrical and optical properties, flexibility and excellent stability, as well as lightweight, radiation resistance and ultra-fatigue resistance, which are particularly needed in future aerospace and military applications. However, to realize the extensive applications of flexible TCFs, it is not only a prerequisite to overcome the mutual restriction between transmittance and conductivity but also to meet the requirement of being able to be prepared on a large area or even on a large scale. This is a knotty problem that has been puzzling researchers in the field of carbon nanomaterials and even in the field of TCF for many years.

The researchers in the "Nanomaterials and Mesoscopic Physics" research group (Group A05) at the Beijing National Laboratory for Condensed Matter Physics and the Institute of Physics (IOP), Chinese Academy of Sciences (CAS), have been committed to the basic investigation on the preparation, properties and potential applications of low-dimensional carbon




Graphic: IOP

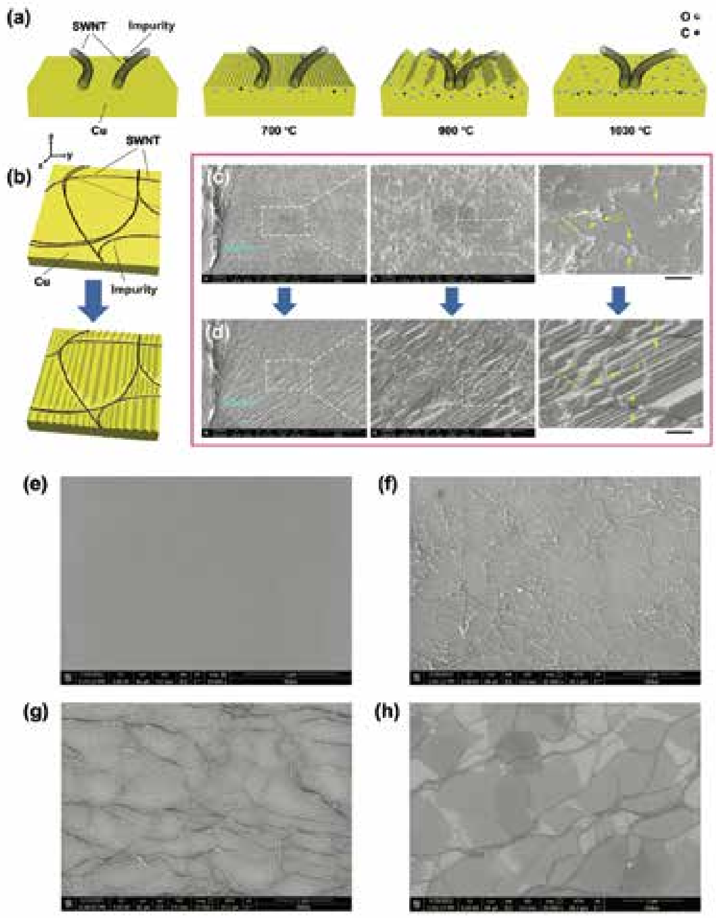
(a-c) Photographs of freestanding G-RSWNT films floating on the water surface with 1m×10cm, A4 size, and A3 size, respectively. (d-g) Transmission spectra, sheet resistances, quality factors, stress-strain curves and Raman spectra of different carbon nanofilms.

nanomaterials and nanostructures for over 30 years, and have achieved a series of important innovative results.

Recently, based on the freestanding transparent conductive carbon nanotube film (CNT TCF) continuously and directly prepared by themselves via the blowing aerosol method (*Advanced Materials*, 32, 2004277, 2020; Patent ZL 201310164499.5, ZL 201811117042.8, PCT Patent US 10,144,647 B2), in view of the above challenging problems, YUE Ying, a PhD candidate at the research group, under the supervision of Prof. ZHOU Weiya, proposed an advanced carbon nanotube network reorganization (CNNR) strategy, designed and developed an innovative facet-driven CNNR (FD-CNNR) technique. Breaking through the bottleneck of mutual restriction between the key properties of carbon nanofilms, they achieved the large-area fabrication and lossless transfer of CNT films. This provides an effective scheme to solve the "stuck neck" problem of large-area flexible TCFs.

Based on the unique mechanism of FD-CNNR technique, this work introduces an interaction between the single-walled carbon nanotube (SWNT) and Cu–O reconfiguration for the first time, which allows the SWNT network to reorganize into a more efficient conductive path. Using this technique, large-area, flexible and freestanding reorganized carbon nanotube TCFs (RNC-TCF) with A3 size or even meter-length have been designed and prepared, including the reorganized SWNT (RSWNT) film and the hybrid film of graphene and reorganized SWNT (G-RSWNT) (the latter has an area of more than 1,200 times that of the existing freestanding hybrid films as reported).

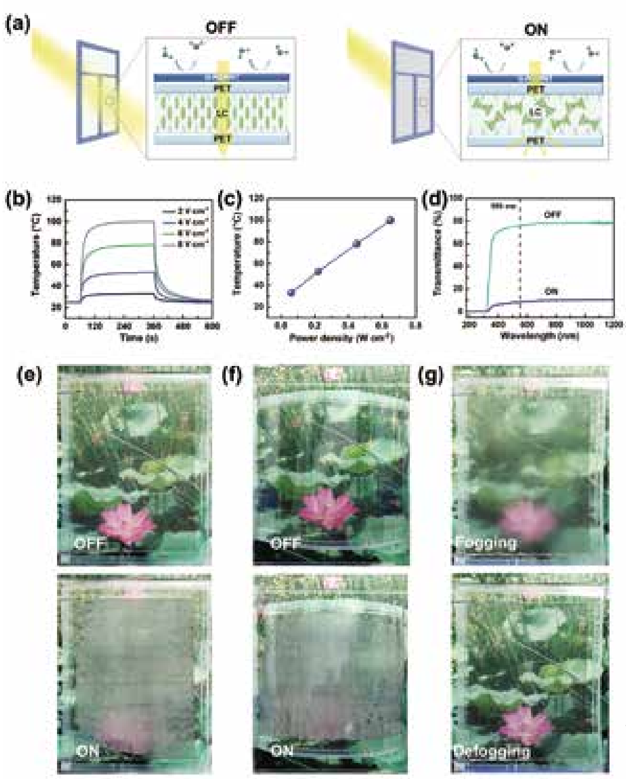
Moreover, the FD-CNNR technique enables these lightweight films to exhibit excellent flexibility, with synergistically enhanced high mechanical strength, outstanding transmittance and conductivity, and significant FOM values. The prepared large-area RNC-TCFs can be freestanding on the water surface and can be transferred to other target substrates without contamination and damage. On the basis of a large-area G-RSWNT TCF and a liquid crystal layer, a new flexible smart window with A4 size has been fabricated, with multi-functions such as fast heating, controllable dimming and defogging. The FD-CNNR technique can not only be extended to large-area or even large-scale preparation of TCFs, but also provide a new idea for the design of TCFs and other functional films.




Graphic: IOP

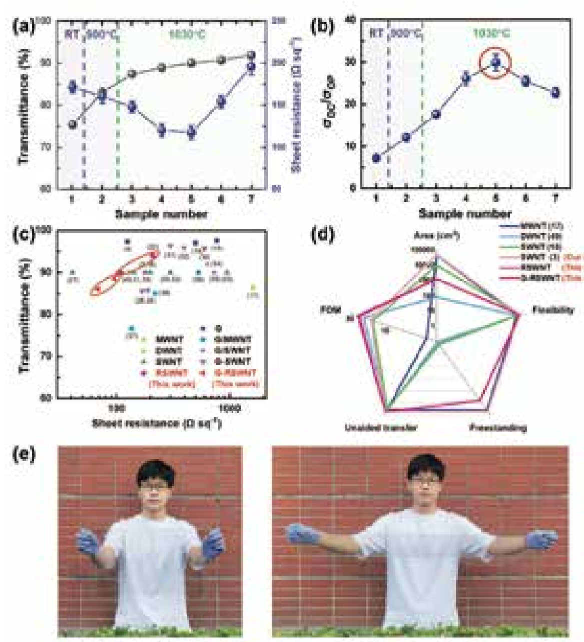
(a) Schematic diagram of the principle of FD-CNNR technique. (b) Schematic diagram of FD-SWNT reorganization mechanism. (c, d) *In situ* SEM images of the reorganization process, with scales of 10 μm, 2 μm, and 500 nm from left to right, respectively. (e-h) SEM images of G, SWNT, RSWNT, and G-RSWNT, with scales of 2 μm.

This work remedies the shortcomings of previous research in the field of large-area graphene-carbon nanotube hybrid films, and is expected to promote the large-scale preparation of large-area, flexible, freestanding, lightweight, and transparent conductive carbon nanofilms and their future applications in the fields of flexible electronics, photovoltaic devices,




Graphic: IOP

(a) Schematic structure and principle of flexible smart window based on G-RSWNT film and liquid crystal layer. (b) Temperature variation of the smart window at different voltage densities. (c) The required power density of the smart window at different steady state temperatures. (d) Transmittance of the smart window in ON/OFF state. (e, f) Transparency change of smart window by voltage regulation at room temperature 25°C, spreading and bending states. (g) Defogging test at 20°C with smart window operating temperature of 28°C.




Graphic: IOP

(a, b) Parameter optimization of SWNT reorganization process. (c) Comparison of sheet resistance and transmittance of the present work with other reported carbon nanofilms. (d) Comparison of multiple properties of the present work with other reported carbon nanofilms. (e) Photographs of G-RSWNT TCF with A3 size and 1m×10cm transferred to PET substrate.



optical engineering, artificial intelligence, modern architecture, transportation, and even aerospace.

The relevant research results have been filed for Chinese invention patent applications. The research paper entitled "Large-Area Flexible Carbon Nanofilms with Synergistically Enhanced Transmittance and Conductivity Prepared by Reorganizing Single-Walled Carbon Nanotube Networks" has been published in the journal *Advanced Materials*. Drs. XIA Xiaogang and LI Shaoqing, PhD candidates ZHANG Di, WANG Pengyu, WU Xin, ZHANG Yuejuan, MEI Jie, and LI Mingming have participated in this research work, with active cooperation and support from Profs. LIU Huaping, ZHANG Xiao, and WEI Xiaojun, and Senior Engineer WANG Yanchun of this group. Here, the authors would like to express their special thanks to and deep memory of Prof. XIE Sishen, late academician of CAS and founder of the research group.

This study was supported by the Ministry of Science and Technology of China, the National Natural Science Foundation of China, and CAS.

Contact:
ZHOU Weiya
Institute of Physics, Chinese Academy of Sciences
Email: wyzhou@iphy.ac.cn

(Source: IOP)