Supporting Information

1.Materials

Citric acid (CA, \geq 99.5%) and dicyandiamide (\geq 98%) were purchased from

China National Pharmaceutical Group Corporation Co., LTD. (Taiyuan, China). L-Arginine (L-Arg) was purchased from China Pharma Group Chemical Reagents Co., LTD. (Shanghai, China). DAF-FM DA (NO fluorescent probe), Cell Counting KIT-8 (CCK-8 Kit), Phosphate Buffered Saline (PBS), Mitochondrial Membrane Potential Detection Kit (JC-1), Nitric oxide detection kit, and Paraformaldehyde (4%) were purchased from Biyuntian Biotechnology Co., LTD. (Shanghai, China). Calcein

Acetoxymethyl Ester (Calcein-AM, ≥96%) and Propidium Iodide (PI, ≥94%), and

ONOO⁻ Fluorescence Detection Kit were purchased from Shanghai Best Biological Co., LTD., China. Fetal Bovine Serum (FBS) was purchased from Hangzhou Tianhang Biochemical Co., LTD., China. High Glucose Culture Medium (DMEM) and Fluorescein Isothiocyanate (FITC) were purchased from Shanghai McLean Biochemical Co., LTD. The 0.25% Trypsin-EDTA solution was purchased from MeilunBio[®]. Human malignant melanoma cells (A-375) and Dylight488 Sheep anti-rabbit IgG were purchased from Bobo Biotechnology Co., LTD (Wuhan, China), and Human Dermal Fibroblasts (HDF) were from Wuhan Saios Biotechnology Co., Ltd. HSP90 Rabbit mAb was purchased from Huidan Biotechnology Co., LTD (Hangzhou, China). Normal goat serum (stock solution) for blocking and Triton X-100 were purchased from Solarbio[®] Science & Technology Co., Ltd (Beijing, China). Hydrogen peroxide 3% purchased from Nanjing Reagent (Nanjing, China).

2. Instruments and characterization

The morphology of the sample was characterized by high-resolution transmission electron microscopy (HR-TEM, JEOL 2100 F, Nippon Electronics, Japan). The surface functional groups of the sample were analyzed using Fourier transform infrared spectroscopy (FTIR, Tensor 27, Bruker Optik, Germany). The absorption spectrum was determined by a UV-vis spectrophotometer (Metash UV-8000A, 190 nm-1200 nm, China). Cell imaging was performed using a fluorescence microscope (Leica Microsystems, Leica DM 3000, China). The cell viability (OD value) was assessed by the CCK-8 method using a microplate reader (Thermo Scientific, Multiskan FC, China). The elemental composition of the sample was analyzed by X-ray photoelectron spectroscopy (XPS, Thermo Scientific ESCALAB 250Xi, USA). In the photothermal experiments, an 808 nm CW laser (MDL-III-

808 nm, CNI, China) and a 730 nm CW laser (MW-GX-730 nm, Laser, China) were used as the irradiated sources, and the distance between the light source position and the aqueous surface of the quartz cuvette was set to 2.5 cm when it could provide the highest power density of 2.5 W/cm². The temperature changes of all samples were recorded with an infrared thermal camera (225 s, Fotric, China).

3. Photothermal conversion efficiency

The photothermal conversion efficiency of Arg-sCNDs aqueous solution was determined using the method proposed by Roper.¹ Initially, 2 mL of the Arg-sCNDs solution (2.5 mg/mL) was placed in a quartz cuvette, and the initial temperature was recorded at room temperature. The solution was then irradiated for 6 min using an 808 nm (730 nm) laser at a power density of 2.5 W/cm². Immediately after irradiation, the laser was turned off, and the solution was allowed to cool naturally back to room temperature. The temperature change of the solution was monitored using an infrared thermal camera. Simultaneously, the temperature of 2 mL deionized water, used as a control, was recorded under identical conditions. The photothermal conversion efficiency (η) of the Arg-sCNDs was calculated using formula (1).

$$\eta = \frac{hA(T_{max} - T_{surr}) - Q_{Dis}}{I(1 - 10^{-A_{\lambda}})}$$
(1)

Among them, T_{surr} and T_{max} represent the ambient temperature and the maximum temperature, respectively. A is the surface area of the colorimetric dish irradiated by the laser. The absorbance A_{λ} of Arg-sCNDs aqueous solution (2.5 mg/mL) at 808 nm (730 nm) is 1.118 (2.762). The incident laser power density (I) at 808 nm (730 nm) is 2.5 W/cm². Q_{Dis} is the heat absorbed by the quartz cuvette itself, and the value measured at 808 nm (730 nm) is 0.036 W (0.044 W) under the same conditions, respectively. Q_{Dis} was obtained in an independent experiment and the specific calculation process was added in the Fig. S4. Therefore, hA in formula (1) is unknown. Then, formulas (2), (3) are introduced to obtain hA.

$$\theta = \frac{T - T_{surr}}{T_{max} - T_{surr}}$$
(2)
$$\tau_s = \frac{m_D c_D}{hA}$$
(3)

Where, m_D denotes the mass of the solution (2 g), c_D is the specific heat capacity of water (value of 4.2 J/g·°C), and τ_s denotes the system time constant. According to formula (4), we can determine the time constant (τ_s).

$$t = -\tau_s(ln\theta) \tag{4}$$

According to formula (3), hA can be determined.



Fig. S1 (a) Absorption spectra and (b) photothermal heating effect (808 nm, 2.5 W/cm²) of Arg-sCNDs in PBS after 0, 2, 7 days.

The stability of Arg-sCNDs dissolved in PBS solution for different times was systematically investigated. For a short-term storage time (2 days), the absorbance and photothermal heating performance maintained stable with negligible changes. However, for a longer period (7 days), significant loss of photothermal properties and attenuation of absorption were observed (Figs. S1a~b). In consideration of the synthesis and storage condition of Arg-sCNDs (solid powder at 4 °C, 60% RH),^{2, 3} "freshly prepared prior to use" was suggested for the use.



Fig S2 (a) UV absorption spectra of Arg-sCNDs (2.5 mg/mL) and Arg-sCNDs after 30 days (2.5 mg/mL). (b) Temperature changes in Arg-sCNDs and Arg-sCNDs after 30 days (2.5 mg/mL) under different power settings of 808/730 nm lasers (2.5 W/cm², 6 min).

The lyophilized Arg-sCNDs powder was stored at 4 °C with 60% RH. After 30 days, the powder was redispersed in Deionized water (2.5 mg/mL), and absorbance spectra were compared with freshly prepared samples (Figs. S2a~b). At the critical NIR photothermal waveband (around 660 nm, the absorbance decreased only 6.7%

after 30 days, confirming minimal degradation of the photothermal active ingredient (Fig. S2a). The $\triangle T_{730/808 \text{ nm}}$ of Arg-sCNDs solution decreased to 29/26 °C from 35.2/28.6 °C under 730/808 nm irradiation (Fig. S2b), indicating the photothermal heating performance was little affected. These results proved the stability of Arg-sCNDs powder under suitable storage conditions (4 °C, Humidity 60% RH).



Fig. S3 Fluorescence spectra of Arg-sCNDs.



Fig. S4 The negative natural logarithm of the cooling time of deionized water versus the temperature gradient after irradiation.

 Q_{Dis} is calculated according to formula (5):

$$Q_{Dis} = h_0 A (T_{w,max} - T_{w,surr})$$
⁽⁵⁾

 h_0A is calculated using DI water as the sample, and the formula is consistent with formula (3) in the main text. The τ_s of DI water at 730 nm and 808 nm are 284.08 s

and 322.84 s (Fig. S4), respectively. $T_{w,max}$ and $T_{w,surr}$ are the maximum and ambient temperatures of DI water during the cooling process after laser is turned off, respectively.



Fig. S5 The absorption spectra of FITC, Arg-sCNDs and Arg-sCNDs-FITC.

The Arg-sCNDs-FITC were prepared as follows:

5 mg FITC was dissolved in 2 mL deionized water. It was then continuously stirred at room temperature and gradually mixed with 10 mL of Arg-sCNDs solution. The whole solution continued to be stirred overnight in the dark to prevent the dye from fading. Finally, the unbound FITC was removed by dialysis with deionized water (critical molecular weight = 3500 Da) in the dark for 72 h, and the Arg-sCNDs-FITC solution was obtained. The UV-VIS-NIR spectrum of the Arg-sCNDs-FITC showd a peak of 483 nm (Fig. S5), indicating that the FITC was successfully labeled the Arg-sCNDs.

References

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- 3. D. Li, D. Han, S. N. Qu, L. Liu, P. T. Jing, D. Zhou, W. Y. Ji, X. Y. Wang, T. F. Zhang and D. Z. Shen, *Light-Sci. Appl.*, 2016, **5**, 8.