Electronic Supplementary Information

A novel signal-on fluorometric sensor based on metal ions-mediated carbon dots for formaldehyde determination and lysosome-targeted bioimaging

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The fluorescence quantum yield measurement

The fluorescence QY of CDs was estimated by the equation[1]:

$$QY_L = QY_R\,[(I_L \times A_R)/(I_R \times A_L)]\,[(N_L)/(N_R)]^2$$

where the subscripts *L* refers to CDs and *R* is the reference, *QY* is the quantum yield (Quinine sulfate in 0.1M H_2SO_4 solution was chosen as reference, the standard *QY* of which is 54% at 360 nm excitation), *I* refers to the integrated intensity of the emission, *N* represents the refractive index of the medium, *A* is the absorbance at corresponding excitation wavelength.

MTT viability assay

Human uterine cancer cell (HeLa) was seeded into 96-well plates with Dulbecco's modified Eagle's medium (DMEM) for 24 h at a density of 10^4 cells/150 μL in an incubator (37 °C, 5% CO_2). And the culture medium was replaced by DMEM containing NSCDs and Ag-M-NSCDs, respectively, with different concentrations (0, 0.01, 0.05, 0.1, 0.25, 0.5, 0.8, 1.0 mg/mL) for another 24 h. Then, the cells were washed by 20 mL PBS buffer (pH 7.4) and incubated with MTT solution (5 mg/mL) for 4 h. After that, the culture medium was removed, followed by the addition of 150 μL DMSO and shaken for 10 min at room temperature. The optical density (*OD*) was measured by a microplate reader (ELx800, Biotek, USA) at 490 nm. The cell viability was estimated according to the following equation:

$$\text{Cell viability (\%)} = (OD_{\text{treated}}/\,OD_{\text{control}}) \times 100\%$$

where OD_{treated} was obtained in the presence of NSCDs/Ag-M-NSCDs, and OD_{control} was obtained in the absence of NSCDs/Ag-M-NSCDs.

Cellular imaging analysis

The HeLa was first cultured in DMEM containing 10% fetal bovine serum (FBS) and 1% penicillin/streptomycin at 37 ℃ in 5% CO_2 condition. Before the imaging experiment, all the cells were allowed to adhere to a confocal dish for 12 h. For the fluorescence imaging experiment of Ag-M-NSCDs and formaldehyde (FA), the cells were incubated with 0.1 mg/mL Ag-M-NSCDs for 6 h and washed with a fresh DMEM. Then, 500 μM FA was added for further incubation of 1 h. For the localization experiment, the HeLa cells were incubated with 0.1 mg/mL Ag-M-NSCDs for 6 h and washed three times with PBS buffer. Then 75 nM Lyso-Traker Cys Deep Red was added in serum-free medium and co-cultured with the cells for 1 h. Finally, the residual probe was

washed three times before imaging under a TCS SP8 (Leica, Germany) confocal microscopy system (*E*x = 405 nm; blue channel, *E*m = 420-460 nm; green channel, *E*m = 520-560 nm; red channel, *E*x = 561 nm, *E*m = 575-630 nm for Lyso-Traker Cys Deep Red).

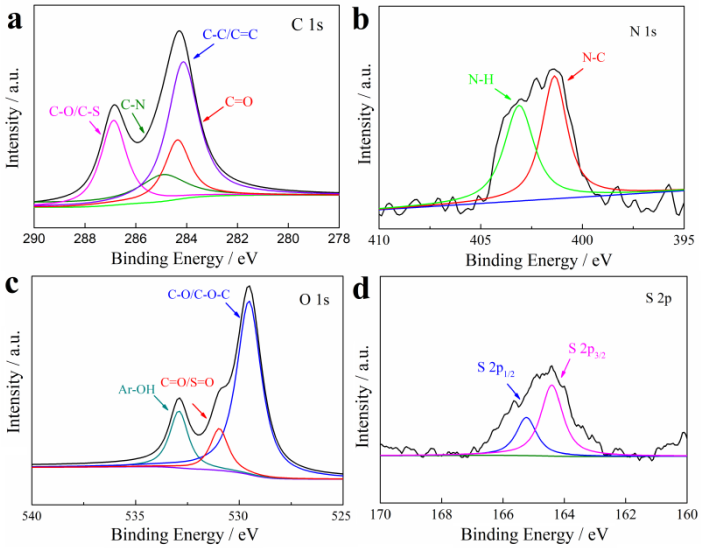

Fig. S1 The high-resolution XPS spectra of (**a**)C 1s, (**b**)N 1s, (**c**) O 1s and (**d**)S 2p in NSCDs.

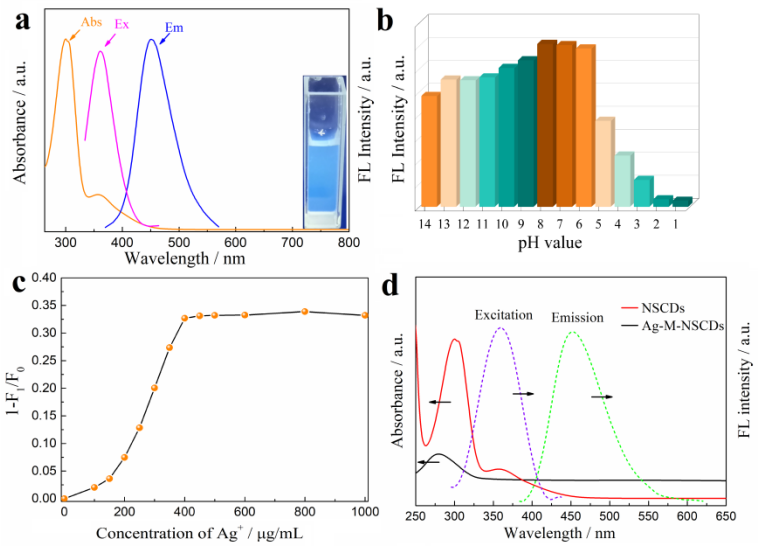

Fig. S2 (**a**) The UV–vis absorption spectrum, fluorescence excitation and emission spectra of NSCDs. The inset is photograph of NSCDs under UV lamp at 365 nm. (**b**)Fluorescence intensities of NSCDs versus different pHs. (**c**) The influence of Ag^{+} concentration on the quenching efficiency of Ag-M-NSCDs. (**d**) The UV-vis absorption spectra of NSCDs, Ag-M-NSCDs and $AgNO_3$ solution, fluorescence excitation and emission spectra of Ag-M-NSCDs.

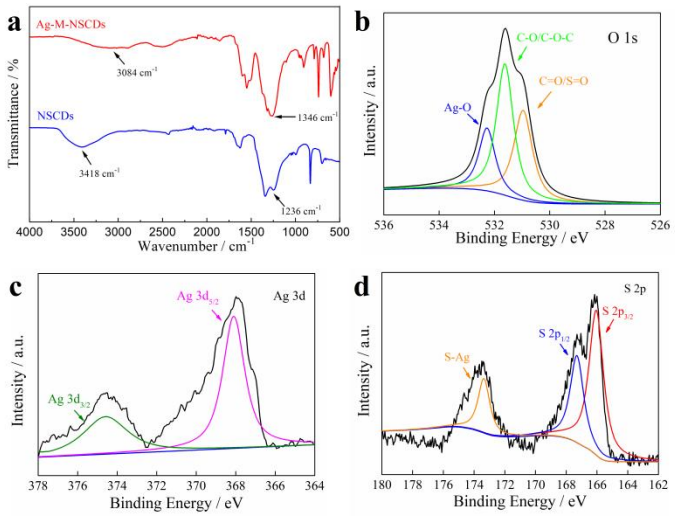

Fig. S3 (**a**) The FT-IR spectra of NSCDs and Ag-M-NSCDs, the high-resolution XPS spectra of (**b**) O 1s, (**c**) Ag 3d and (**d**) S 2p in Ag-M-NSCDs.

Table S1 Determinations of FA in tap water

Sample	Add/μM	Measured/μM		Recovery/%	RSD/%
		HPLC-MS	This work		
1	0.00	6.23	6.75	/	3.22
2	50.00	47.89	48.21	96.42	2.58
3	250.00	257.56	259.25	103.70	5.83
4	500.00	532.22	529.10	105.82	5.11

Table S2 Determinations of FA in PBS

Sample	Add/μM	Measured/μM		Recovery/%	RSD/%
		HPLC-MS	This work		
1	0.00	/	/	/	3.31
2	50.00	44.75	45.89	91.78	4.65
3	250.00	248.21	255.48	102.19	5.47
4	500.00	483.34	524.35	104.87	4.77

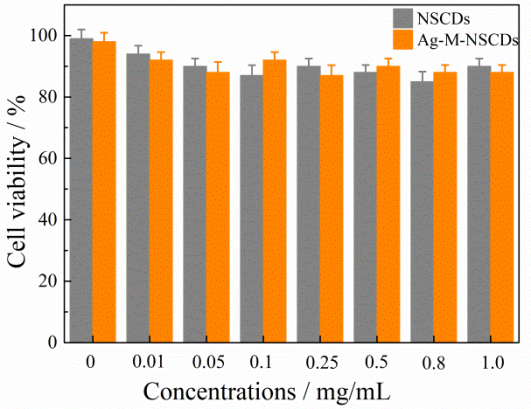

Fig. S4 Cell viability of HeLa cells after incubated with a series concentrations of NSCDs and Ag-M-NSCDs, respectively.

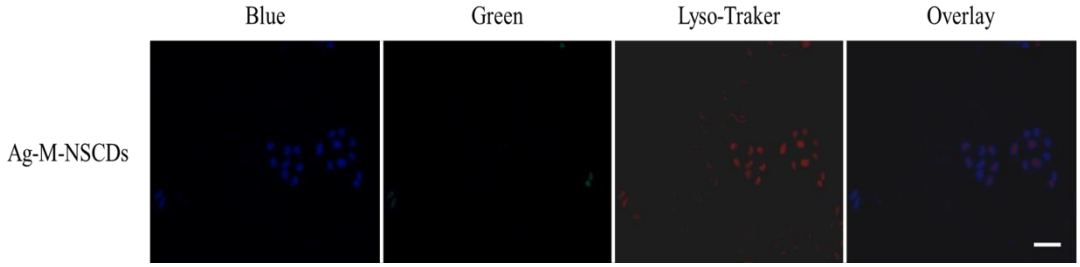

Fig. S5 Fluorescence images of HeLa cells incubated with Ag-M-NSCDs and the Lyso-Traker Cys Deep Red.

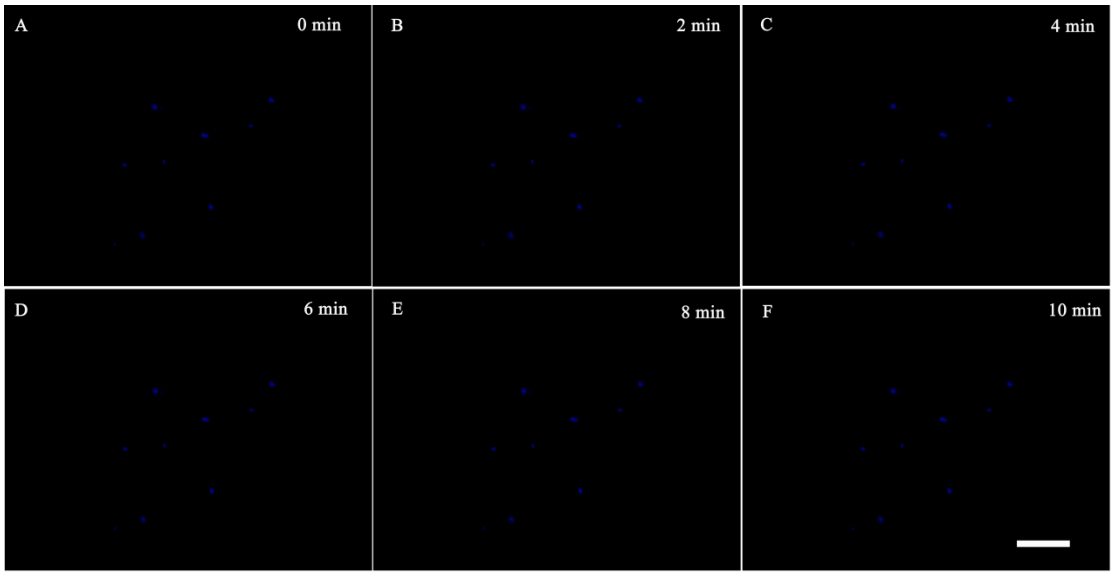

Fig. S6 Fluorescence images of HeLa cells after incubation in the medium containing Ag-M-NSCDs and then with a continuous scan of 10 min.

References

[1] X.Y. Chai, H. He, H.H. Fan, X.H. Kang and X.P. Song, A hydrothermal-carbonization process for simultaneously production of sugars, graphene quantum dots, and porous carbon from sugarcane bagasse, Bioresour. Technol., 2019, 282, 142-147.