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## A Co-crystallization Induced Surface Modification Strategy with Cyanuric Acid Modulates the Bandgap Emission of Carbon Dots

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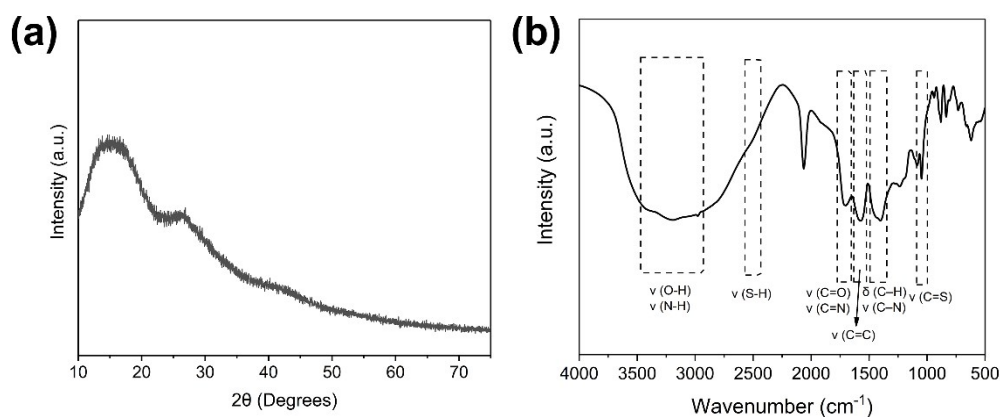
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### Experimental details on fluorescence and phosphorescence intensity measurements

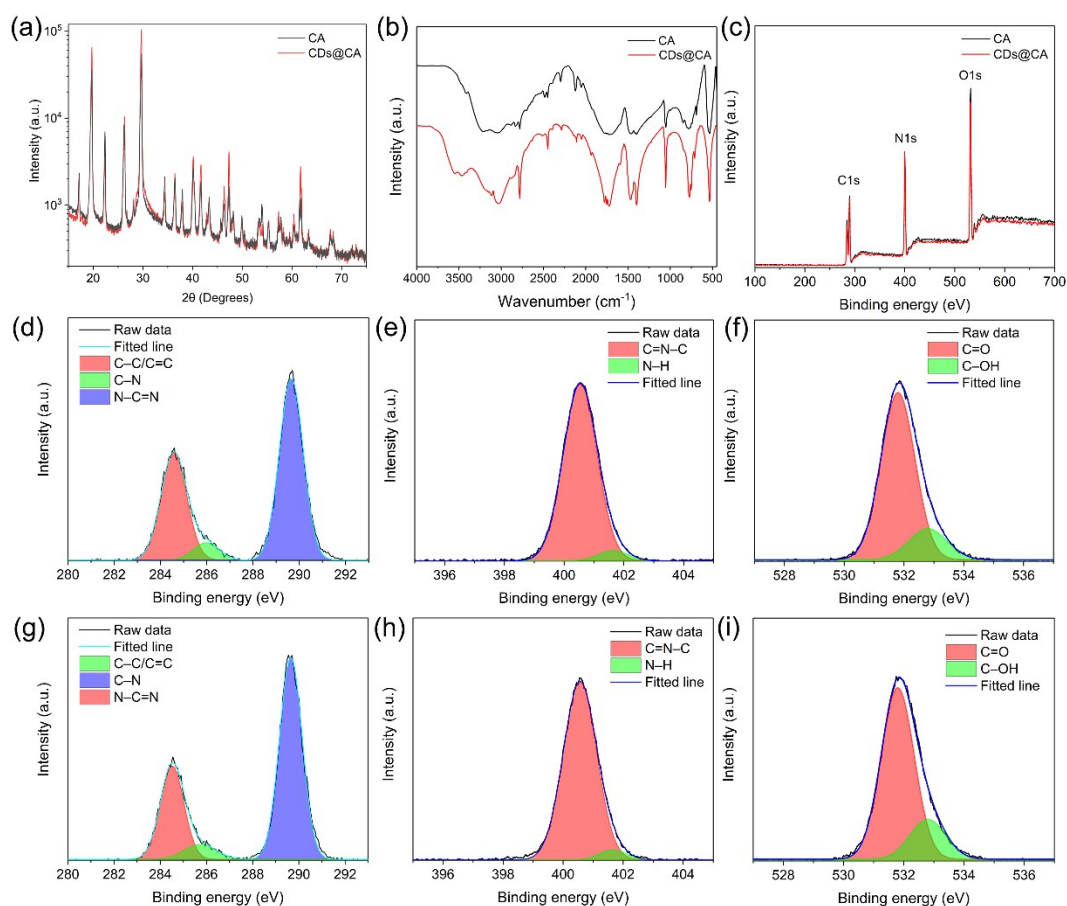
During the heating-cooling cycles of the CDs@CA powder, we fixed the position of the excitation UV light and the optical fiber of the NOVA-EX spectrometer, using a fiber spectrometer to measure the intensity of the fluorescence, and using a camera to take pictures of the fluorescence. In the phosphorescence measurement, we use the camera's high-speed continuous shooting function (10 photos / second) for continuous shooting during the process of turning off the excitation light. The second photo after the excitation light was turned off was selected as the phosphorescent photo of the sample. The instantaneous phosphorescence intensity at that temperature was obtained by photometric analysis of the images using ImageJ software.

**Table S1** PL QYs of CDs, CDs/CA in different solvents and of the solid CDs@CA crystals.

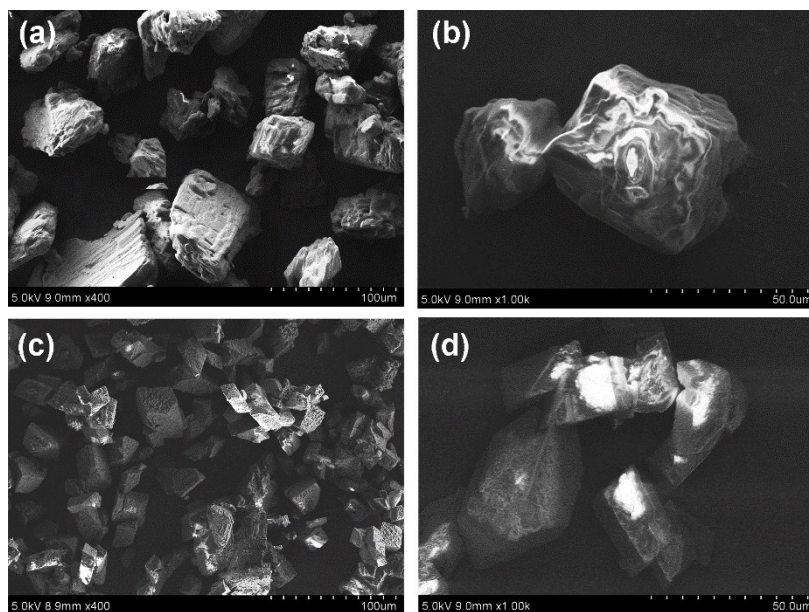
| Excitation wavelength | CDs in H <sub>2</sub> O | CDs in DMSO | CDs/CA in H <sub>2</sub> O | CDs/CA in DMSO | CDs@CA crystals |
|-----------------------|-------------------------|-------------|----------------------------|----------------|-----------------|
| 360 nm                | 10%                     | 9.7%        | 4.5%                       | 4.6%           | 23%             |
| 420 nm                | 12.8%                   | 12.5%       | 24%                        | 57%            | 62%             |



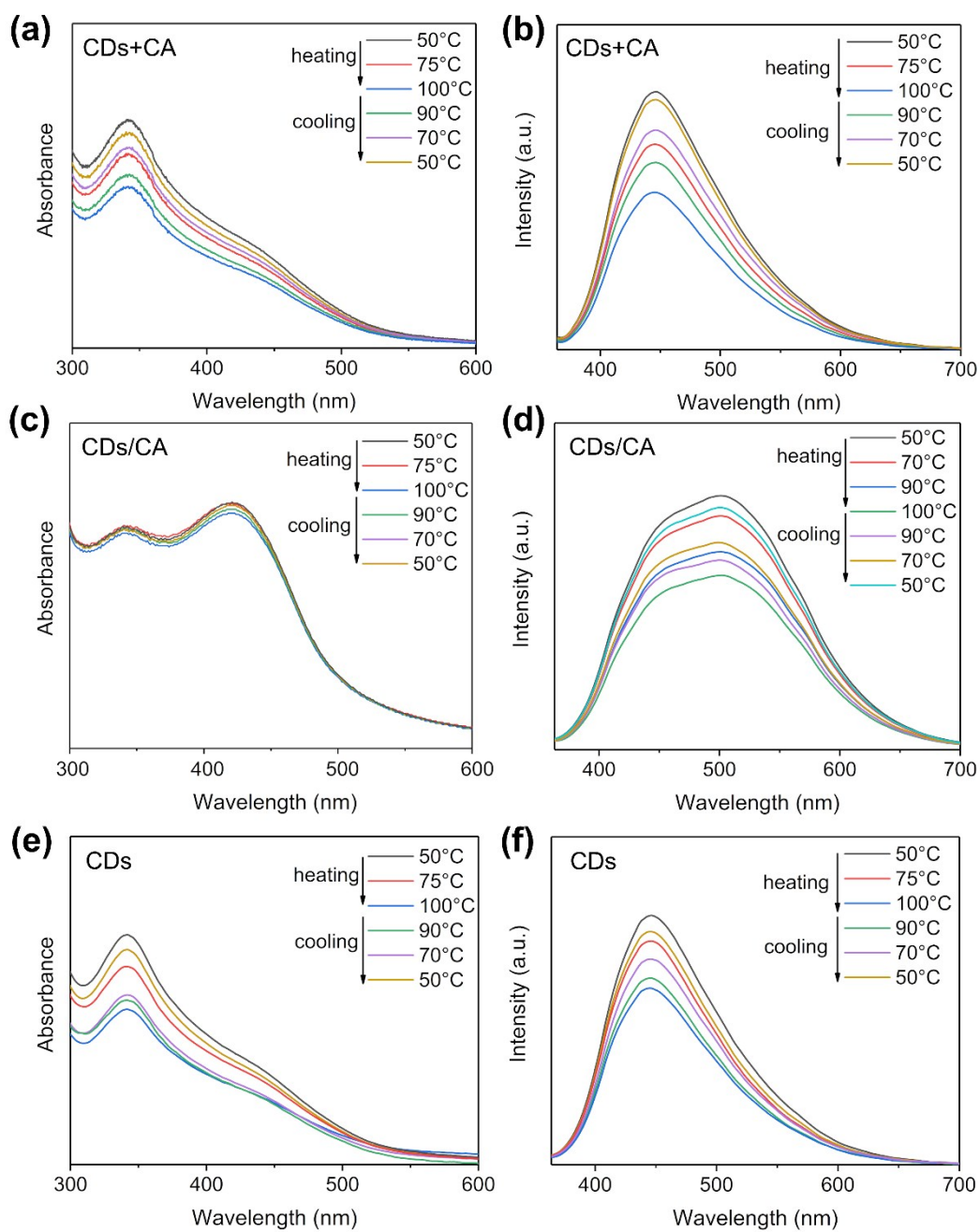
**Figure S1** (a) XRD pattern and (b) FTIR spectrum of CDs.



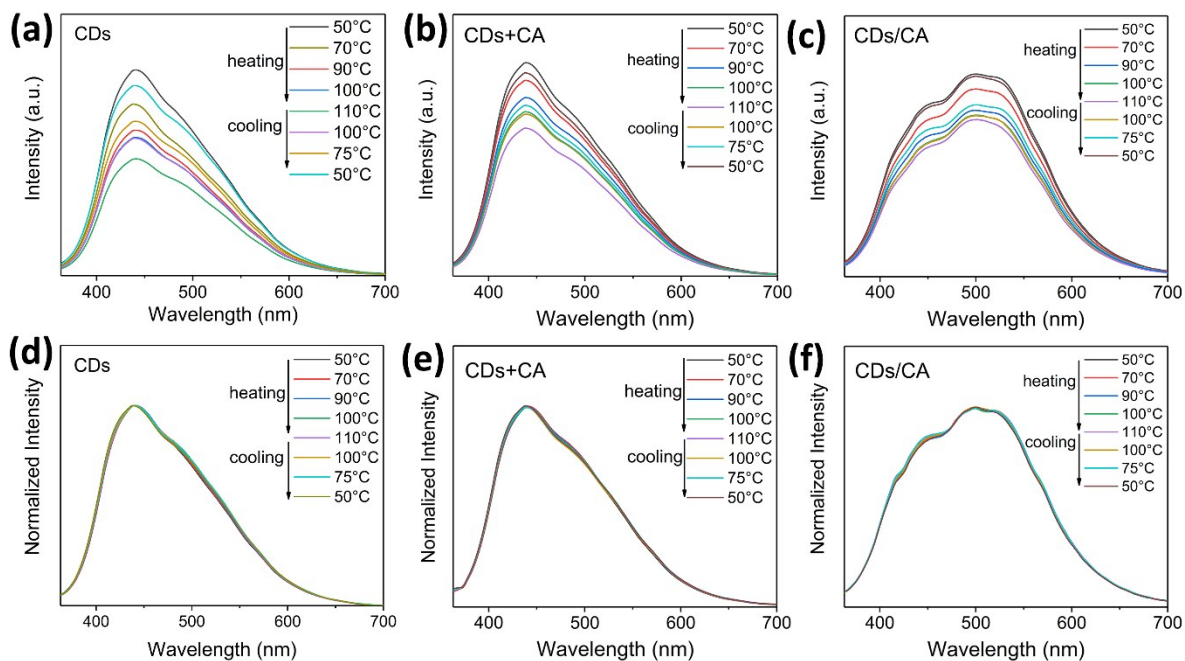
**Figure S2** (a) XRD spectra, (b) FTIR spectra and (c) full survey XPS spectra of the CA crystals and CDs@CA crystals. (d) C1s, (e) N1s and (f) O1s XPS spectra of the CA crystals. (g) C1s, (h) N1s and (i) O1s XPS spectra of the CDs@CA crystals.



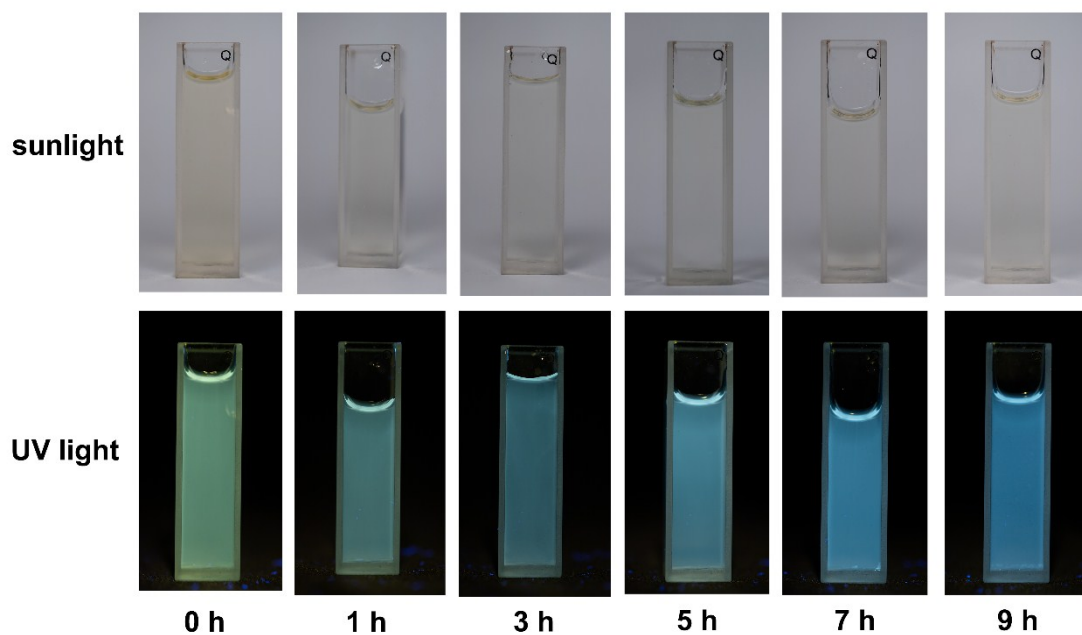
**Figure S3** SEM images of (a, b) CA crystals and (c, d) CDs@CA crystals.



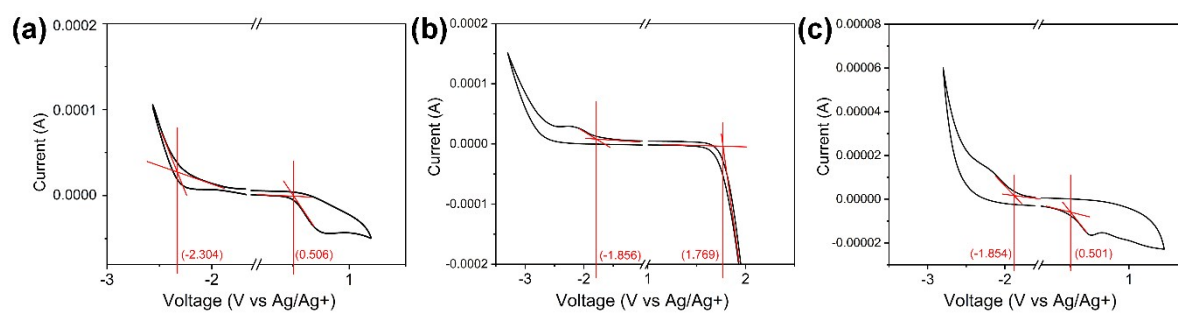
**Figure S4** (a, c, e) UV-Vis absorption and (b, d, f) PL spectra taken during the heating/cooling cycles of (a, b) CDs+CA, (c, d) CDs/CA, and (e, f) CDs aqueous solutions.



**Figure S5** PL spectra of (a) CDs (b) CDs+CA and (c) CDs/CA in DMSO solutions, taken over heating/cooling cycles, as indicated. (d), (e) and (f) are normalized versions of (a), (b) and (c), respectively.



**Figure S6** Photographs of CDs/CA aqueous solutions after hydrothermal heating at 160 °C for different periods of time, as indicated.



**Figure S7** Cyclic voltammograms of (a) CDs, (b) CA, and (c) CDs/CA in DMF solution.