Electronic Supplementary Information

Preparation and optical properties of highly luminescent colloidal single-layer carbon nitride

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The choice of the temperature regime for the synthesis of bulk carbon nitride (CN). Typically, the melamine is condensed in the temperature range of 520–550 °C. At a temperature higher than 600 °C partial elimination of residual amino groups takes place with the release of ammonia and some increase of the atomic C:N ratio. At a temperature higher than 700 °C gradual destruction of the CN occurs with the products depending on the pyrolysis conditions.

In this work the temperature of 650 °C was chosen for the preparation of the bulk C_3N_4 to be then exfoliated into the luminescent colloidal CN sheets. It was found that complete exfoliation of 1.0 g bulk CN in 20.0 mL of aqueous 1.36 M solution of NEt_4OH occurs after the thermal treatment for 4 h at around 100 °C at constant magnetic stirring as described in the Experimental section. In the same conditions only half that mass of CN produced at 520 or 550 °C can be dispersed with the formation of a much less stable

colloidal solution and the rest of the bulk CN forming a sediment. Optical measurements showed that after dilution to the same concentration, colloidal CN particles produced at 650 °C (CN-650) manifest 7–8 times higher photoluminescence intensity than that of the colloidal CN produced at 550 °C (CN-550). The origins of the enhanced photoluminescence of CN-650 as compared to CN-550 are currently unclear and will state a subject of a separate study. The CN sample produced at 700 °C and than exfoliated in similar conditions also reveals by an order of magnitude lower photoluminescence intensity, most probably due to partial destruction of the heptazine network and formation of carbon contamination that has a light-screening effect. Therefore, the pyrolysis temperature of 650 °C was chosen in the paper as producing the most intense luminescing colloidal solution.

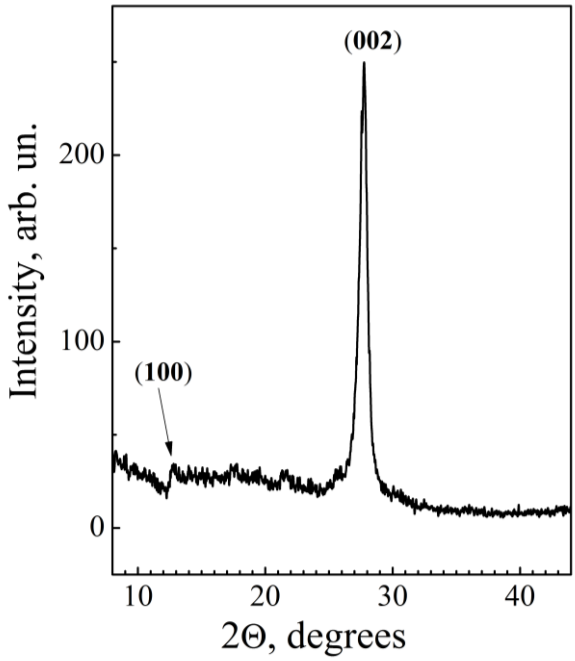
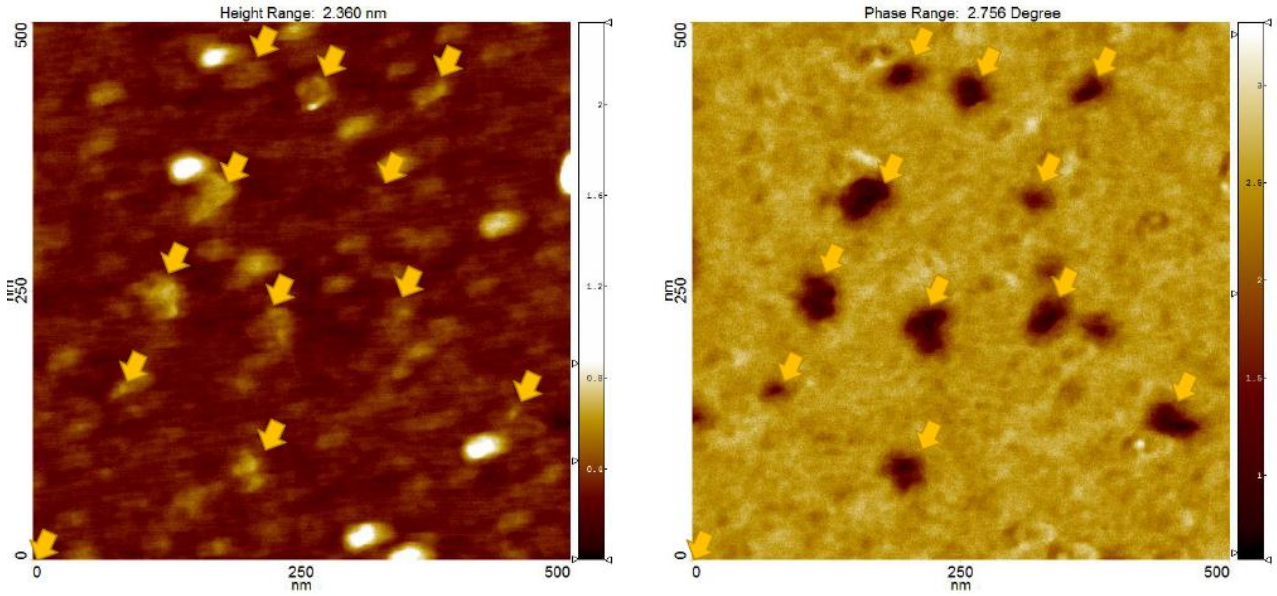
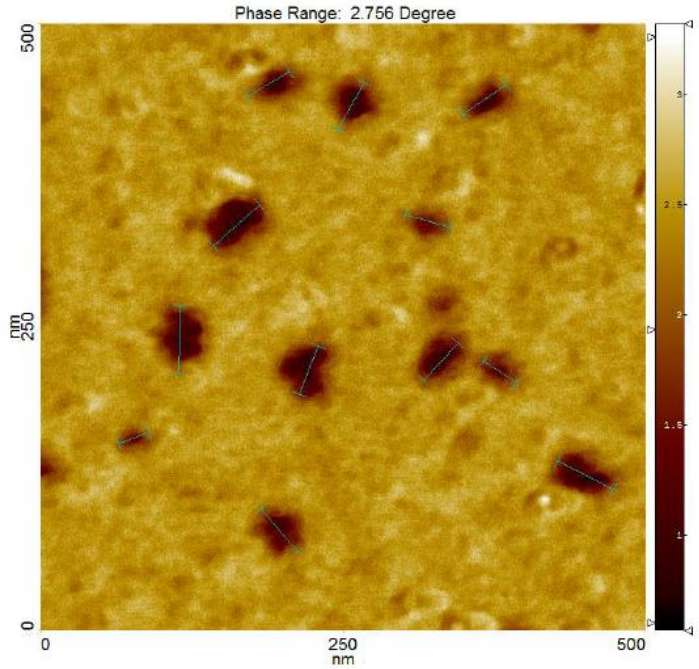


Figure S1. X-ray diffraction pattern of bulk g-C_3N_4. XRD pattern was registered using a Bruker D8 Advance diffractometer with Cu K_α irradiation (1.5418 Å) in a range of 2Θ = 0.4–60° with a scanning rate of 1 grade/min.



a b

Figure S2. Height-contrast (a) and phase-contrast (b) AFM images of an area of mica plate with colloidal single-layer CN particles.



ID	Length [nm]
1	51.1
2	40.0
3	45.2
4	44.4
5	38.2
6	56.4
7	26.5
8	42.4
9	41.7
10	51.6
11	33.1
12	46.5
Minimum	26.5
Maximum	56.4
Mean	**43.1**
Std. Dev.	**8.21**

Figure S3. Phase-contrast AFM image and statistics on the lateral size of colloidal single-layer CN particles.

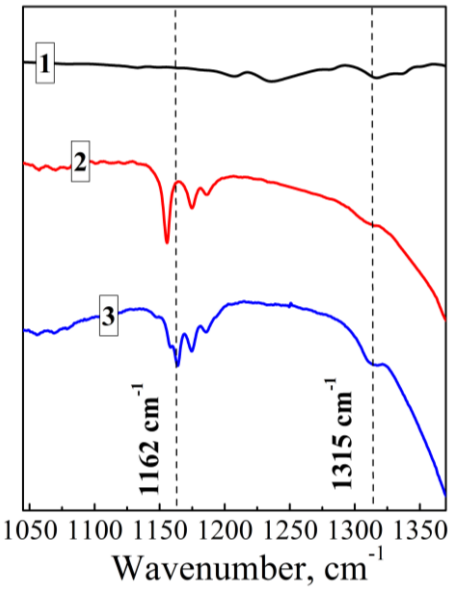


Fig. S4. Fragments of IR spectra of bulk g-CN (curve 1), NEt_4OH (curve 2), and CN exfoliated in NEt_4OH solution (curve 3).

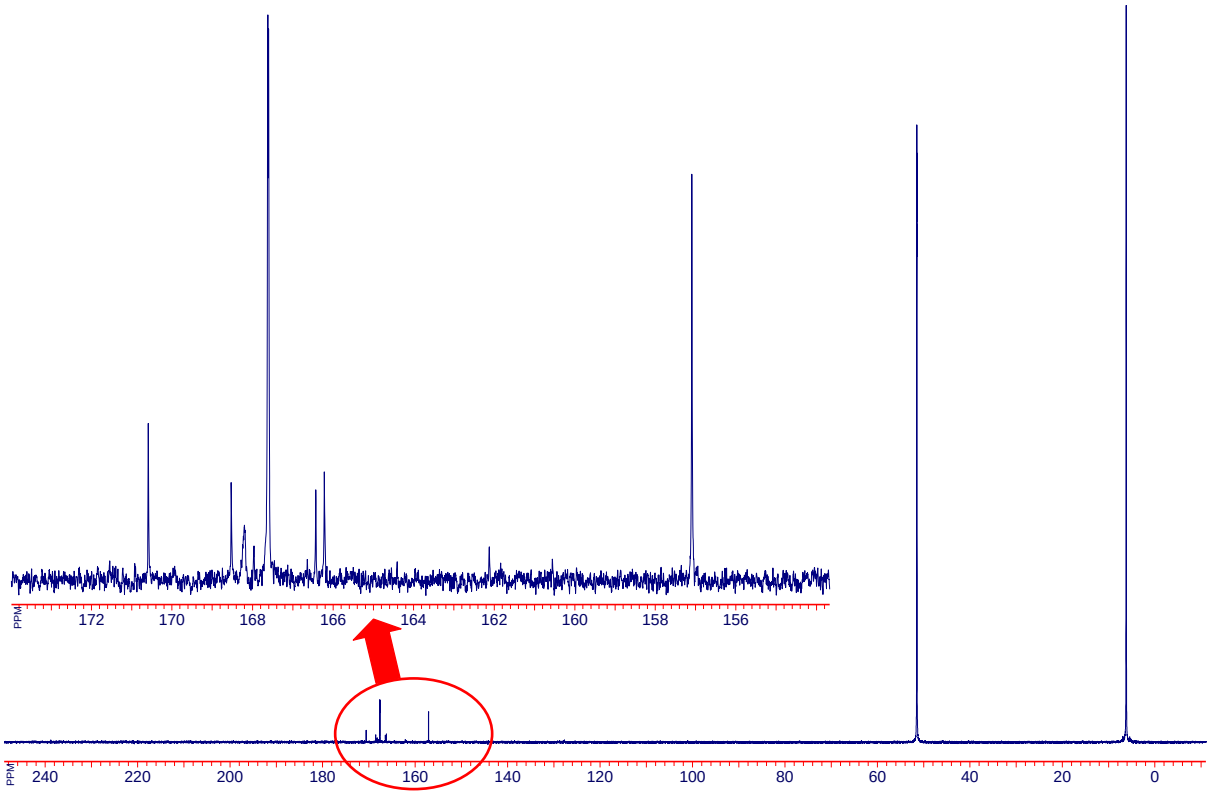


Figure S5. ^{13}C NMR spectrum of colloidal CN.