Supporting Information

Synthesis of Smooth and Bamboo-like Well-Crystalline CN_x Nanotubes with Controllable Nitrogen Concentration (x = 0.05~1.02)

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Table S1: A) The typical morphologies and x values of the as-obtained CN_x products by adding different amounts of reagents.

Adding amount of reagents					x value	Morphology	Figure
$CO(NH_2)_2$	NaN_3	$NaNH_2$	C_2H_5OH	Mg	of CN_x	of NTs	No.
3.10 g	2.00 g	–	–	2.05 g	1.02	Smooth	1
3.10 g	1.10 g	–	–	2.05 g	0.52	Smooth	S2
3.10 g	–	3.00 g	–	2.05 g	1.01	Bamboo-like	3
3.10 g	–	1.65 g	–	2.05 g	0.49	Bamboo-like	S3
3.10 g	–	–	–	2.05 g	0.08	Smooth	S4
3.10 g	–	–	2 mL	2.05 g	0.05	Smooth	S5

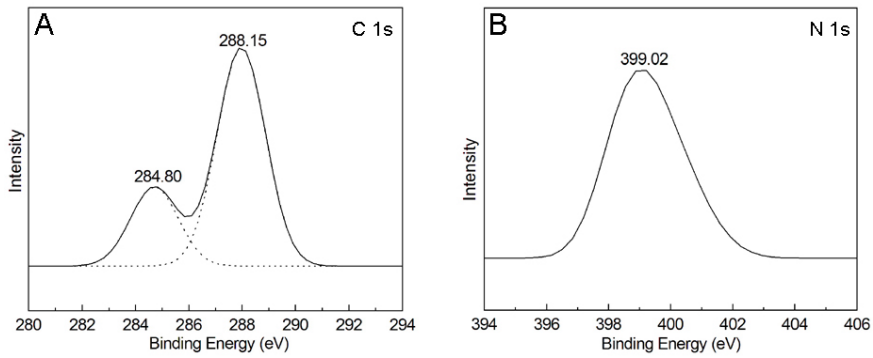

Figure S1. (A) C1s XPS peaks and (B) N1s XPS peaks of the sample obtained by adding 2.00 g NaN_3. XPS measurements indicate that the C1s (Figure S1A) and N1s (Figure S1B) binding energies of the sample are 288.15 and 399.02 eV, respectively, which can be attributed to sp^2 C and sp^2 N in atomic structure of graphite-like CN. The observed C1s and N1s binding energies are very similar to C-N bonds of graphite-like CN (Alba de Sánchez, N.; Carrasco, C.; Prieto, P. *Physica B* **2003**, 337, 318).

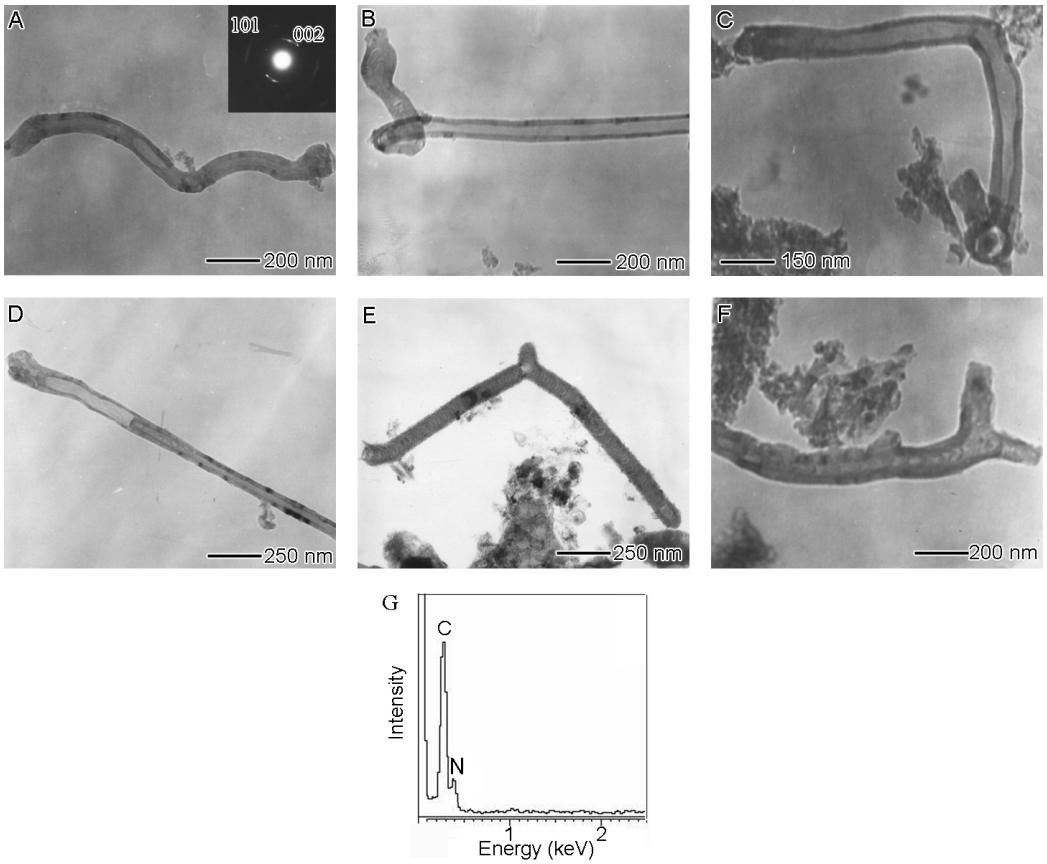

Figure S2. (A)-(C) TEM images and ED pattern of smooth $CN_{0.52}$ NTs with large bending angles synthesized by adding 1.10 g NaN_3. (D) TEM images of subdivided $CN_{0.52}$ NTs synthesized by adding 1.10 g NaN_3. (E), (F) TEM images of Y-structural $CN_{0.52}$ NTs synthesized by adding 1.10 g NaN_3. (G) EDX spectrum of $CN_{0.52}$ NTs synthesized by adding 1.10 g NaN_3. By changing the adding amount of sodium azide, the nitrogen concentration of smooth CN_x NTs (i.e., the value of x) can be continuously adjusted. For example, the $CN_{0.52}$ NTs were also prepared by adding half amount of sodium azide. It was found that the CN_x NTs with lower nitrogen concentration have similar properties with carbon NTs. As examples, many $CN_{0.52}$ NTs with large bending angles (Figure S2A-C) were observed; there also exist some Y-structural (Figure S2D) and subdivided NTs (Figure S3E and F) in the products. The nitrogen concentration was determined by EDX (Figure 2G) and elemental analysis measurements.

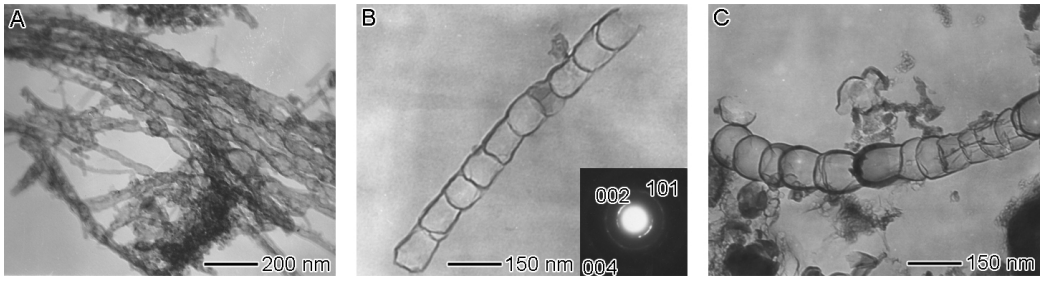

Figure S3. TEM images and ED pattern of bamboo-like $CN_{0.49}$ NTs prepared by adding 1.65 g $NaNH_2$. The nitrogen concentration of bamboo-like CN_x NTs can be continuously adjusted by changing the adding amount of sodium amide. For example, bamboo-like $CN_{0.49}$ NTs were obtained by adding half amount of sodium amide.

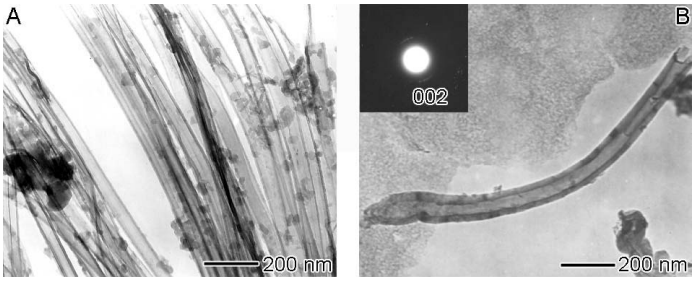

Figure S4. TEM images and ED pattern of smooth $CN_{0.08}$ NTs obtained from the reaction between Mg and $CO(NH_2)_2$. A sealed reaction between urea ($CO(NH_2)_2$) and metal magnesium (Mg) at 550~600 °C could produce smooth CN_x ($x \cong 0.08$) NTs. It indicates that the urea could act as both carbon and nitrogen source in the reactions.

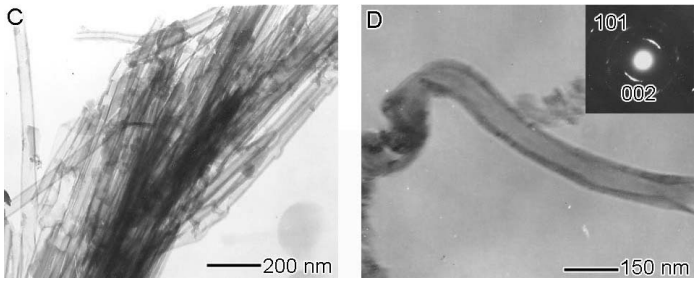

Figure S5. TEM images and ED pattern of smooth $CN_{0.05}$ NTs prepared by adding 2 mL C_2H_5OH. With x value below 0.08, the nitrogen concentration can be controlled by introducing carbon source of ethanol (C_2H_5OH). As a typical case, Figure S5 shows TEM images of smooth $CN_{0.05}$ NTs obtained by introducing 2 mL ethanol.

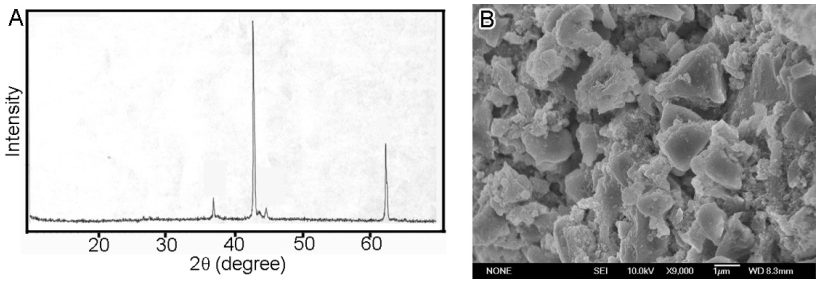

Figure S6. (A) XRD pattern and (B) FE-SEM image of the as-obtained product from the reactions of 2.05 g metal magnesium (Mg), 3.10 g urea ($CO(NH_2)_2$) and 3.40 g sodium azide (NaN_3). The elemental analysis results show that the N/C ratio of the product from the reactions of 2.05 g metal magnesium (Mg), 3.10 g urea ($CO(NH_2)_2$) and 3.40 g sodium azide (NaN_3) is 1.34, which is good agreement with the C_3N_4 stoichiometry. The XRD pattern (Figure S6A) of the product cannot be precisely indexed to any phase of C_3N_4. Some of the diffraction peaks are similar to those of α-C_3N_4 and β-C_3N_4, however, the strongest peaks of these two phases cannot be found in the obtained pattern. Totally, the product is impossible to be graphitic-C_3N_4, since the (002) peak cannot be observed in the pattern. The FE-SEM image (Figure S6B) of the product shows that the product is composed of some sub-microcrystals.

Instrumentation. FE-SEM images and EDX spectra were taken on a JEOL JSM-6700 SEM. TEM images and ED patterns were taken on a Hitachi Model H-800 instrument with a tungsten filament using an accelerating voltage of 200 kV. HRTEM images and ED patterns were recorded on a JEOL-2010 TEM at an acceleration voltage of 200 KV. Element analysis was taken on an ELEMENTAR VARIO EL-III elemental analyzer. XRD patterns were performed on a Mac Science MXPAHF rotating anode X-ray diffractometer with Cu Kα radiation (λ = 1.54178 Å). XPS spectra were collected on an ESCALab MKII X-ray photoelectron spectrometer, using nonmonochromatized Mg-KII X-ray as the excitation source.