Electronic Supplementary Material (ESI) for RSC Advances

Supporting Information

Facile synthesis of carbon nitride micro-/nanoclusters with photocatalytic activity for hydrogen evolution

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Experimental section

Materials. Cyanamide, Celatom FW-14, NH_4HF_2 and Triethaolamine were obtained from Sigma-Aldrich and used without further treatment.

Synthesis. Carbon nitride micro-/nanoclusters were synthesized through a solid "incipient wetness impregnation" method. 1 g of Celatom FW-14 and certain amount of cyanamide were mixed evenly in the mortar and transferred into the crucibles with lid and heated under N_2 atmosphere to the desired temperature (450~650 °C, ramp rate 2.3 °C·min^{-1}) and kept at the temperature for another 4 hours. For the removal of diatom, the resultant yellow powder is treated with 4 M NH_4HF_2 solution with stirring for 24 hours. The dispersion is then filtered; the precipitate is copiously rinsed with deionized water and ethanol. After filtering procedure, the yellow powder is dried under vacuum at 60 °C overnight.

Characterizations. XRD measurements were performed on a D8 Diffractometer from Bruker instruments (Cu Kα radiation, λ = 0.154 nm) equipped with a scintillation counter. N_2 sorption experiments were done with a Quantachrome Autosorb-1 at liquid nitrogen temperature. TEM images were taken on Philips CM200 FEG (Field Emission Gun), operated at an acceleration voltage of 120 kV. SEM measurement was performed on a LEO 1550 Gemini instrument.

Photocatalytic hydrogen evolution on carbon nitride micro-/nanoclusters. In a typical experiment, 50 mg carbon nitride material was placed in the photoreactor (equipped with a magnetic stirrer) then the reactor was evacuated and refilled with argon for five times. Then 38 mL of aqueous TEOA solution (10 vol% TEOA) containing the platinum precursor (Pt metal content: 3% weight of carbon nitride) was added under argon flow. The set-up is equipped with a thermostat and stirring plate. H_2O and TEOA were pretreated before use. H_2O was firstly degassed for 1 h under vacuum in an ultrasonic bath and secondly purged with argon for 1 h. TEOA was purged for 1 h with argon. After the reaction, a sample of the headspace was analyzed with GC for hydrogen content.

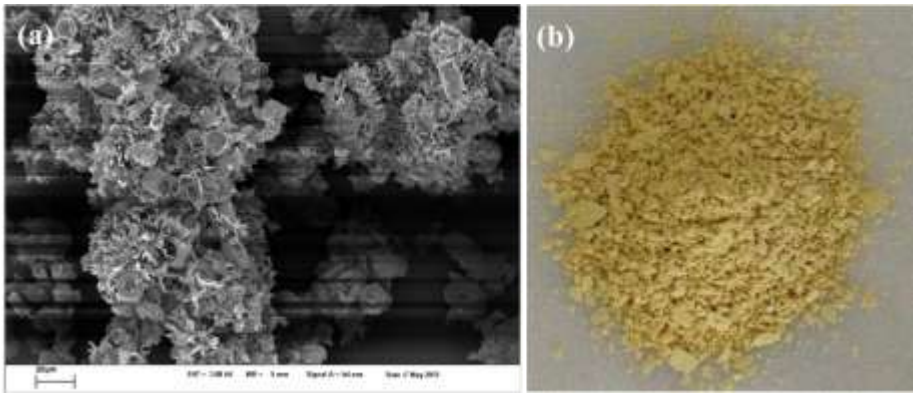

Figure S1. (a) Large area overview of $CNMN_1$-600 material, demonstrating the homogeneous distribution of micro-/nanoclusters. (b) The optical digital image shows the free flowing property of obtained materials.

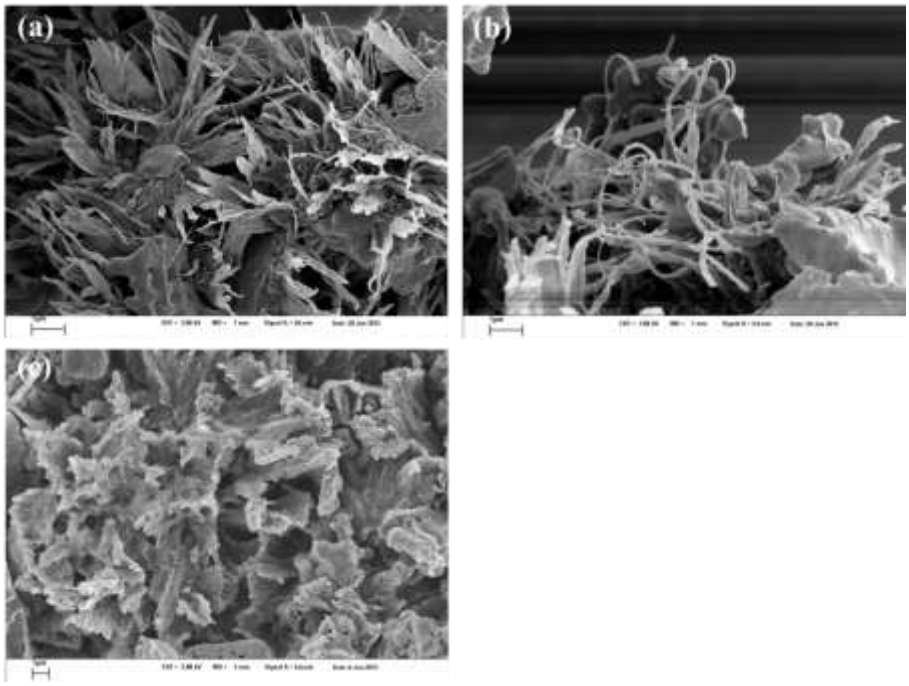

Figure S2. (a) SEM image of micro-/nanoclusters grown from carbon nitride coating on the diatom frustule; (b) SEM image of curved carbon nitride nanowire; (c) SEM image of free standing micro-/nanoclusters after removal of diatom substrate.

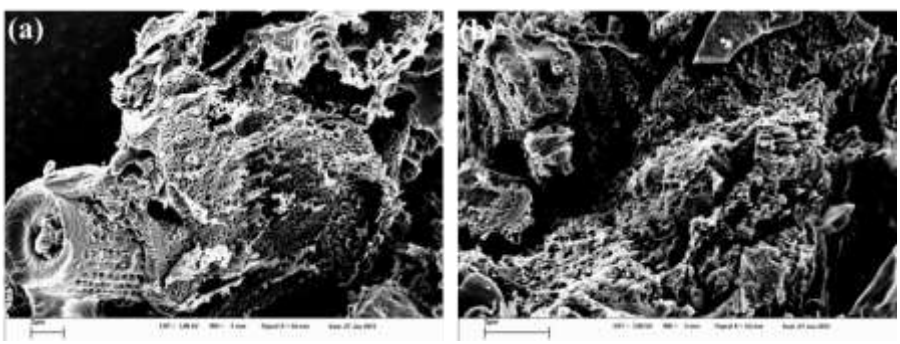

Figure S3. SEM images of $CNMN_2$-600 (a) and $CNMN_3$-600 (b). The high cyanamide content leads to particles intergrown after condensation instead of free flowing. Some porous structures could still be clearly observed from the images.

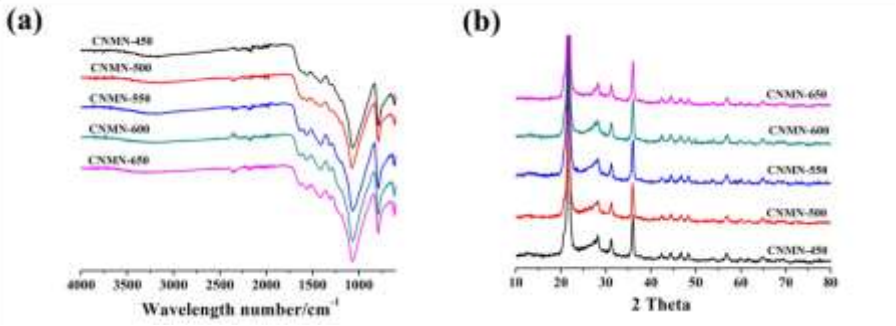

Figure S4. XRD (a) and FTIR (b) of carbon nitride/diatom composites under different calcination temperatures for $CNMN_1$-T.

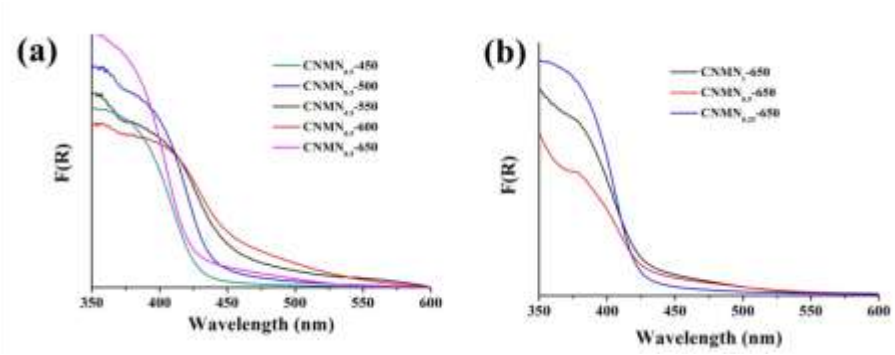

Figure S5. Diffuse reflectance spectra of $CNMN_{0.5}$-T (a) and CNMNx-650 (b).

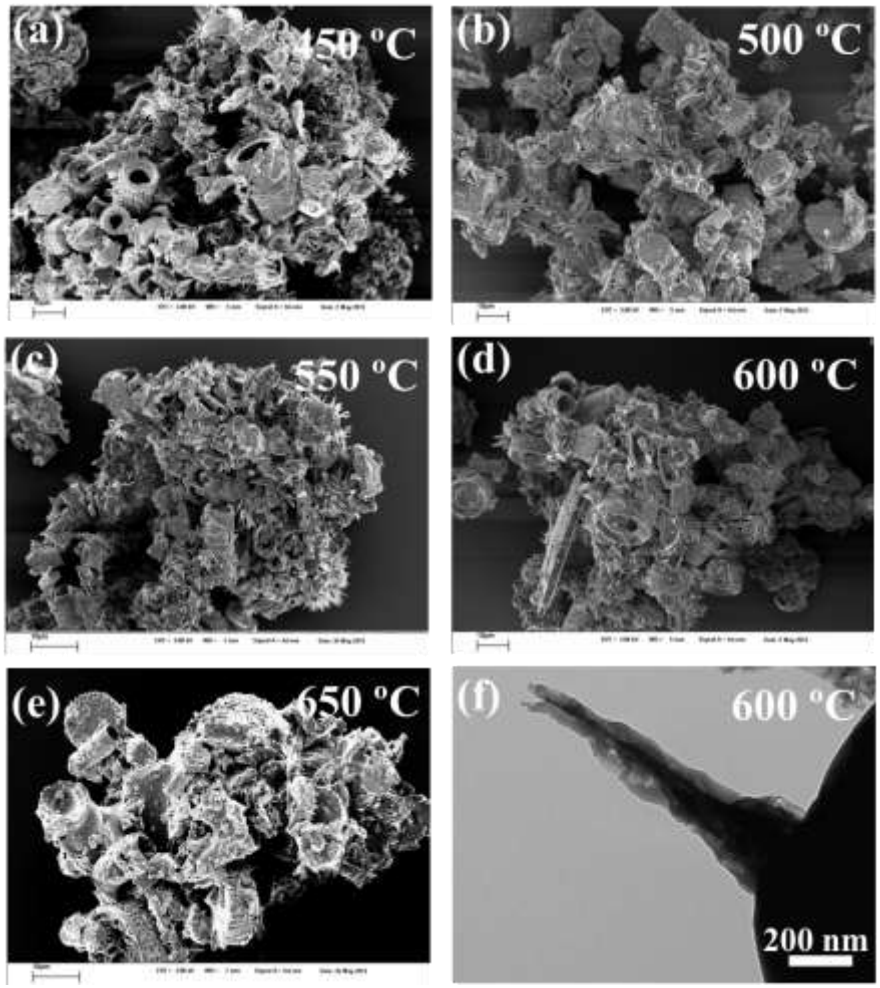

Figure S6. SEM images of $CNMN_{0.25}$-T, demonstrating the nanowire morphology prevails across different condensation temperature (a-d) except 650 ºC condition (e). TEM image of CNMN0.25-600 in (e) shows the nanowire grows outside from the diatom substrate.

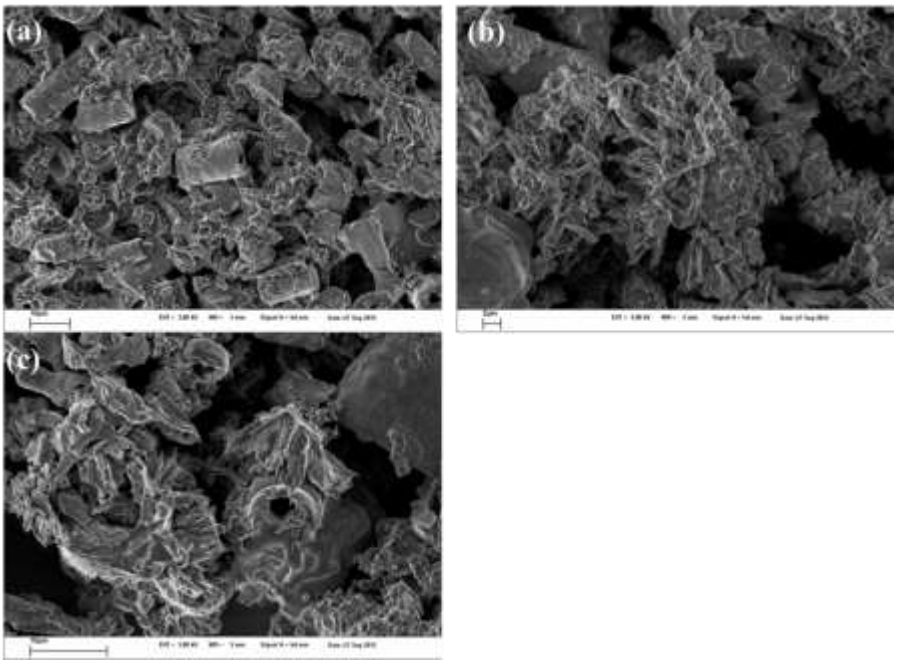

Figure S7. SEM of recycled CNMN. (a) CNMN0.5-600; (b) CNMN0.5-600; CNMN1-600.

	Color	Forward Voltage Vf (V)			Test Current (mA)	Typical Coefficient of Forward Voltage (mV/ºC) Δf/Δj	Power (Typ) (W)	Efficacy (Typ at T_j 25ºC) (lm/W)
		Min	Typ	Max				
BXRA-50C5300-H-00	Cool white	21.9	24.4	26.8	2100	-8 to -24	51	117

Table S1. Electrical characteristics of white LED lamp employed in the photocatalytic reaction.