Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Supporting Information For:

Ammonia Controlled Synthesis of Monodispersed N-Doped Carbon Nanoparticles

Farzana Hanif, Yingcen Liu, Jihong Liu, Liyan Zhang, Hua Lin, Rongwen Lu*, Shufen Zhang

State Key Laboratory of Fine Chemicals, Dalian University of Technology, Dalian 116024, China

Corresponding Author: Rongwen Lu, E-mail: lurw@dlut.edu.cn

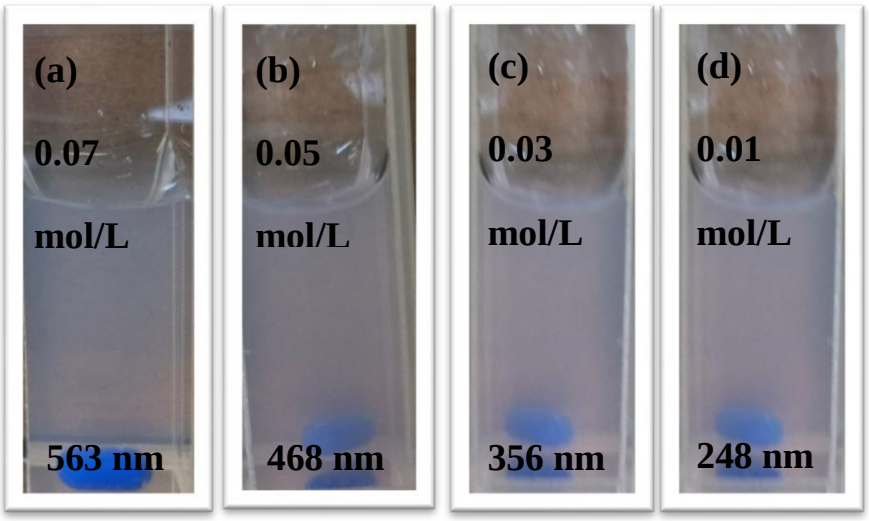

Fig. S1 Photographs of the solution (a-d) with different ammonia concentration, turbidity times (a= 1 min 35 sec, b= 1 min 20 sec, c= 1 min, d= 50 sec,) and different particles size

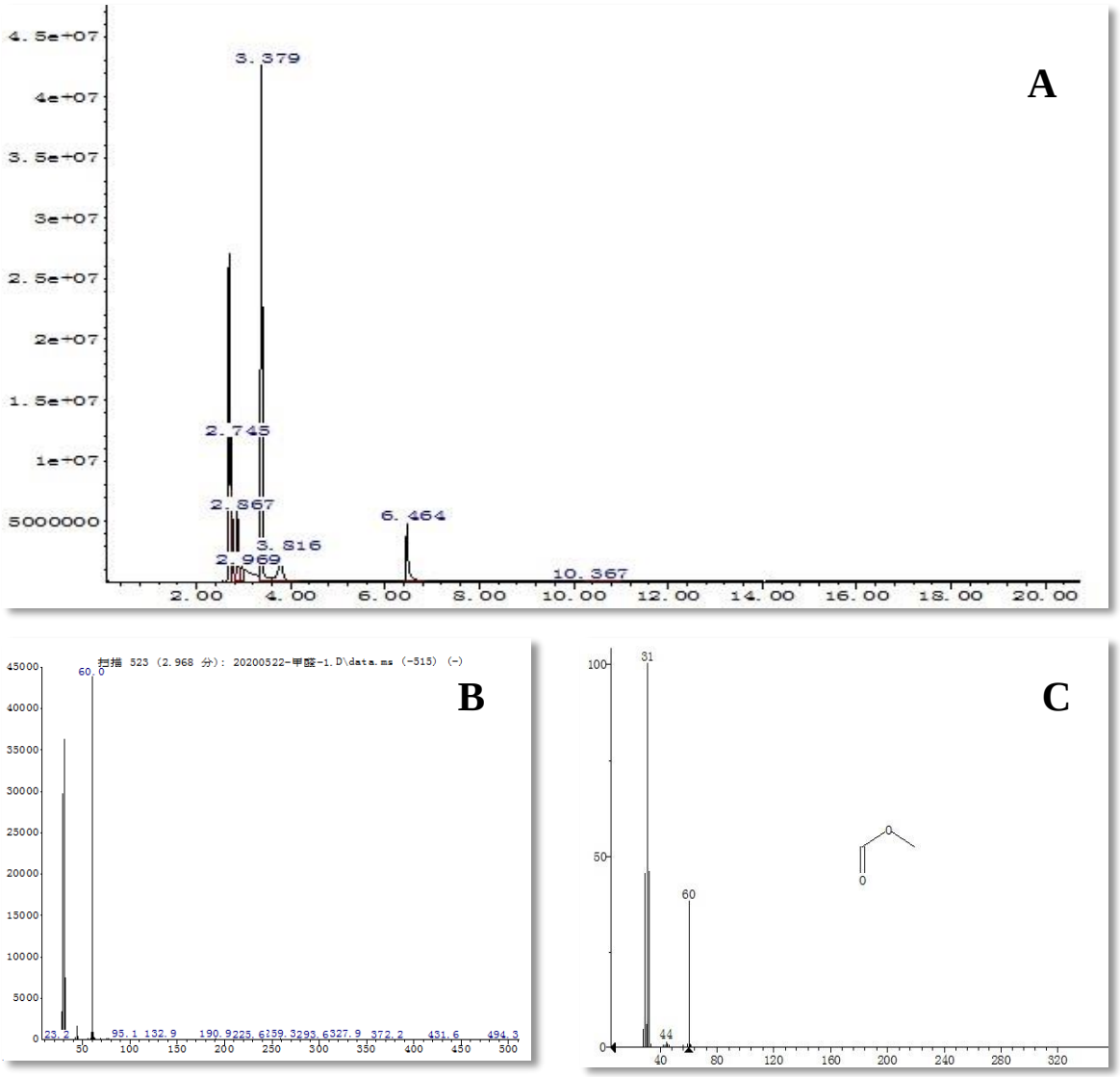

Fig.S2 (A) GC spectrum of HCHO solution, (B) mass spectrum of methyl formate with retention time of 2.96 min (C) magnified mass spectrum of methyl formate.

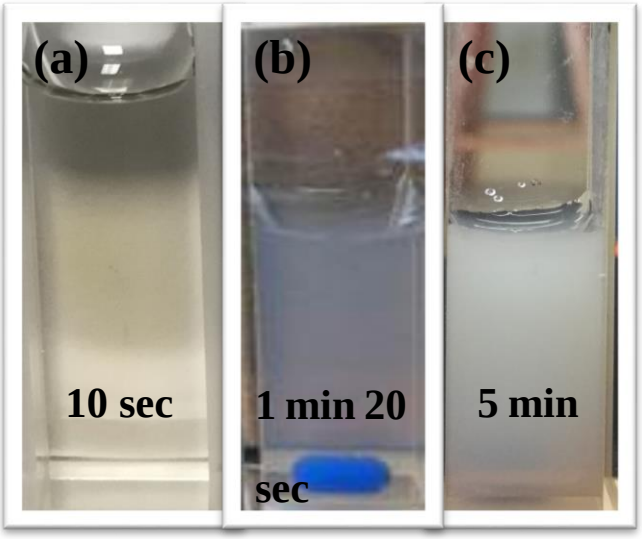

Fig. S3 Photographs of the solutions states changed with time after addition of HCHO (a, 10 sec; b, 1 min 20 sec; c, 5 min). The reaction was conducted in 2 mL solution of *p*-PDA (0.5 mmol), with 1:3 molar ratio of formaldehyde to ammonia.

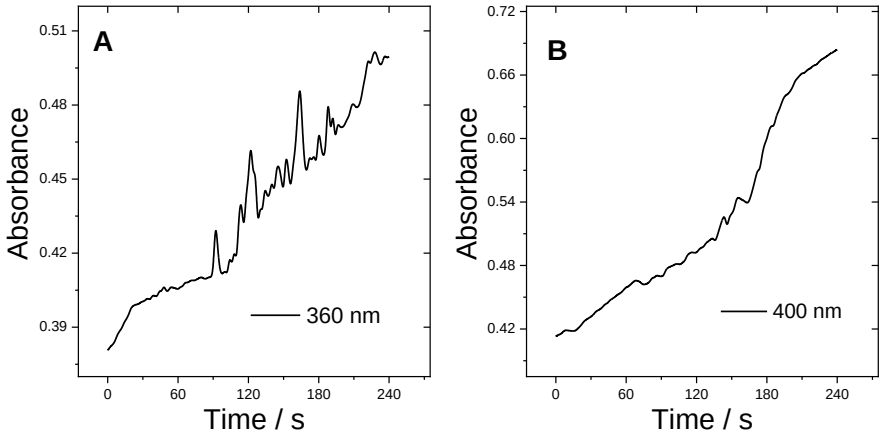

Fig. S4 Time drive graph collected for reaction systems with different ammonia to formaldehyde mole ratios, recorded at different wavelengths (360 nm, 400 nm) showing noise.

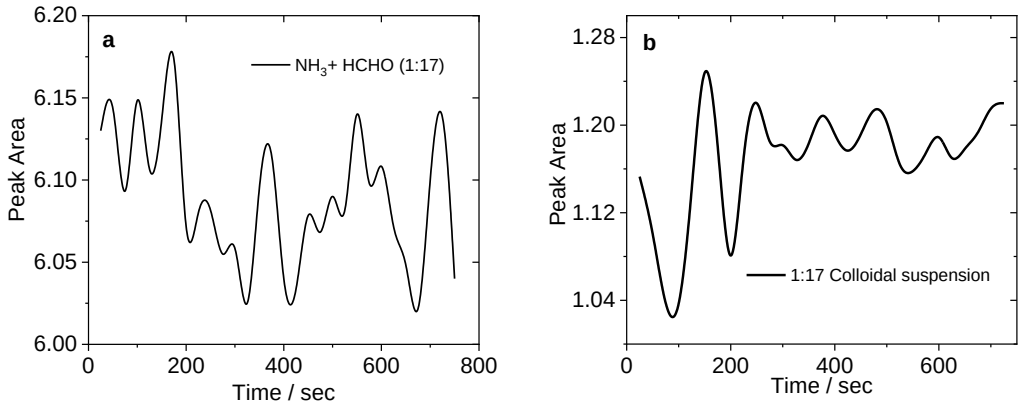

Fig. S5 (a) Relationship between time and integral area of the peak at δ 3.34 for 1,3,5-hexahydrotriaznie from reaction of NH_3+HCHO with molar ratio 1:17. (b) Relationship between time and integral area of the peak for same intermediate at δ 3.34 for colloidal suspension with molar ration 1:17.

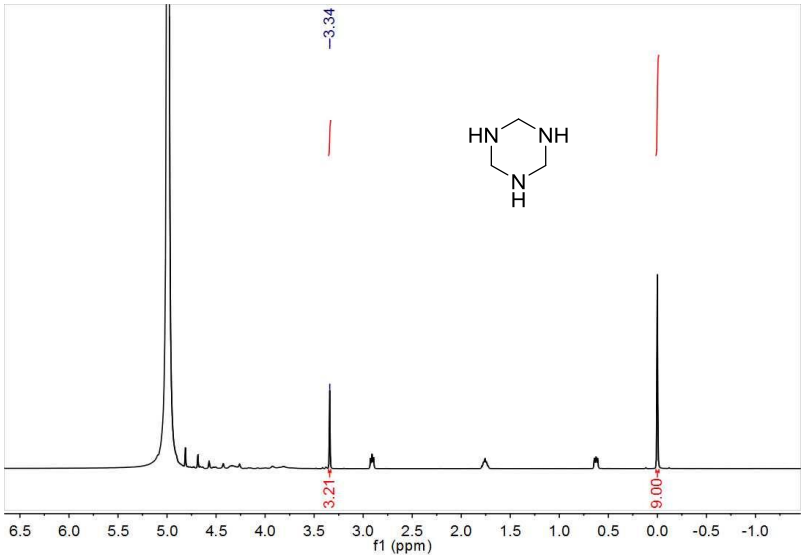

Fig. S6 ^{1}H NMR graph for the ammonia and HCHO reaction with molar ratio 1:17. The peak at δ 3.34 is for three methylene groups in 1,3,5-hexahydrotriazine. This is the only intermediate of formaldehyde and ammonia reaction which appeared in ^{1}H NMR from

starting to end and shows the reversibility trend.

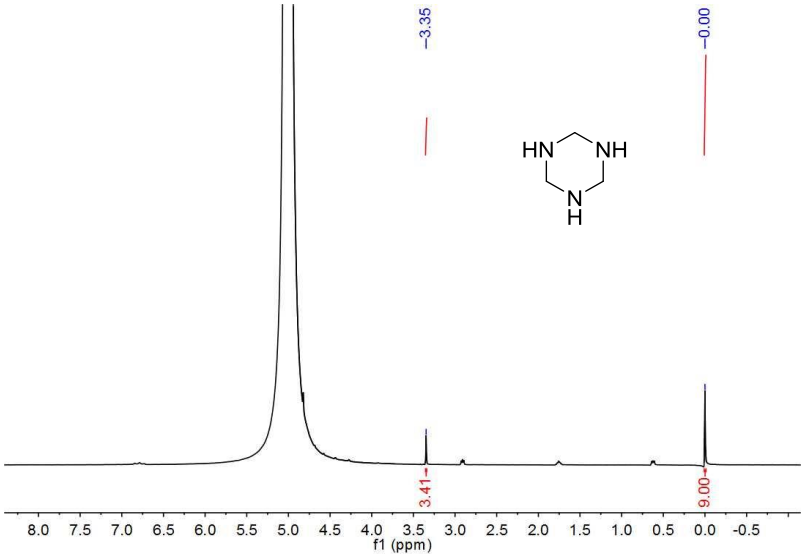

Fig. S7 ^{1}H NMR graph for the PDAs with 1: 17 mole ratio of ammonia to HCHO. The same peak at δ 3.34 is for three methylene groups in 1,3,5-hexahydrotriazine. This is the common and only intermediate peak detected by ^{1}H NMR only for both reactions.

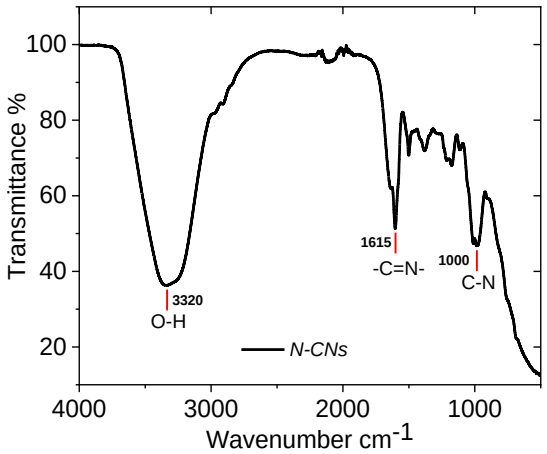

Fig. S8 FT-IR spectrum of PDAs.

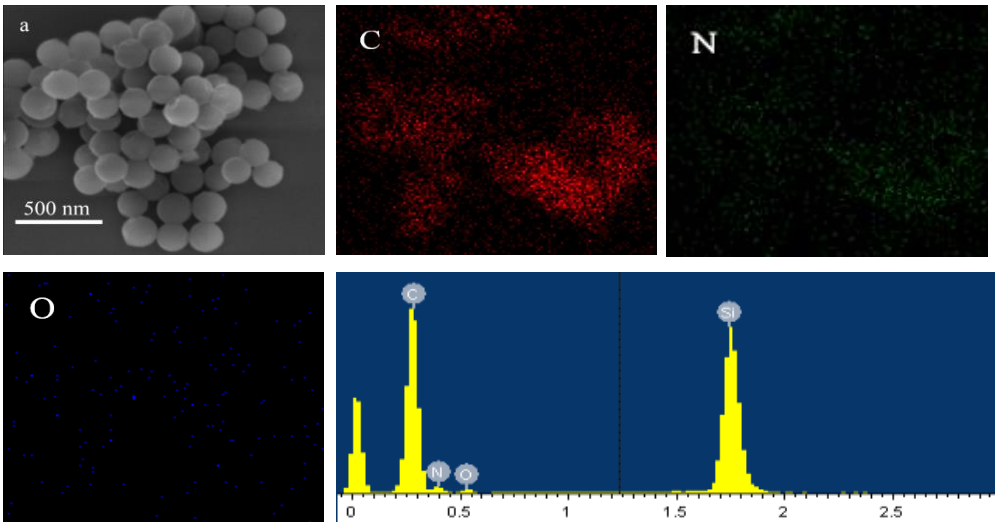

Fig. S9 SEM-EDS for elemental mapping of N-CNs.

Table. S1 Percentage of C, N, and O contents distribution in N-CNs.

Elements	Weight %	Atomic %
C	71.29	78.25
N	14.87	13.99
O	3.56	2.94
Si	10.28	4.82
Total	100.00	100.00

Silicon was the base of testing sample in SEM-EDS analysis.

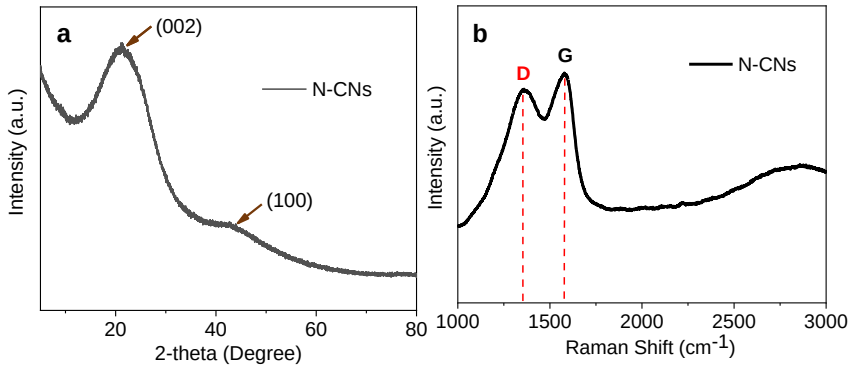

Fig. S10 XRD pattern of N-CNs (a), Raman spectrum of N-CNs (b).

Table. S2 Elemental composition by XPS

Entry	Name	Atomic%
1	C 1s	82.59
2	N 1s	14.79
3	O 1s	2.61

CO_2 gas adsorption analysis

Fig. S11 (a) and (b) displayed the pure adsorption isotherms of CO_2, CH_4, and N_2 at 273 and 298 K, at 1 bar. The uptake of CO_2, CH_4 and N_2 are displayed in Table S3.

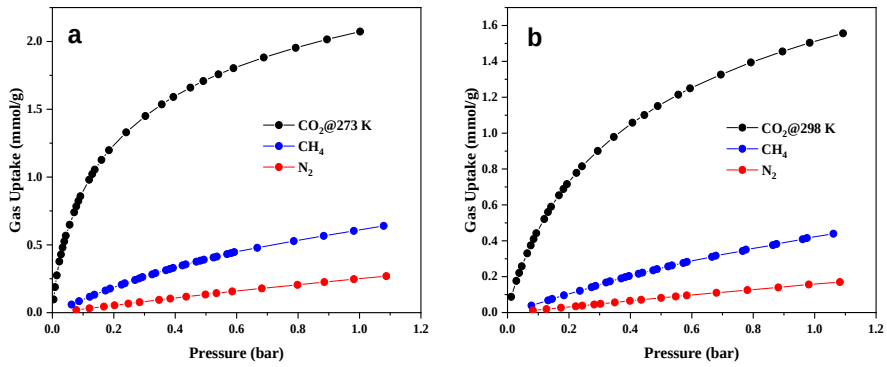

Fig. S11 Adsorption isotherms comparison of CO_2, CH_4, and N_2 at 273 K (a), and 298 K (b).

Table. S3 Gas uptake capacities at different temperatures.

Gas uptake	273 K	298 K
CO_2	2.5	1.6
CH_4	0.8	0.6
N_2	0.40	0.24

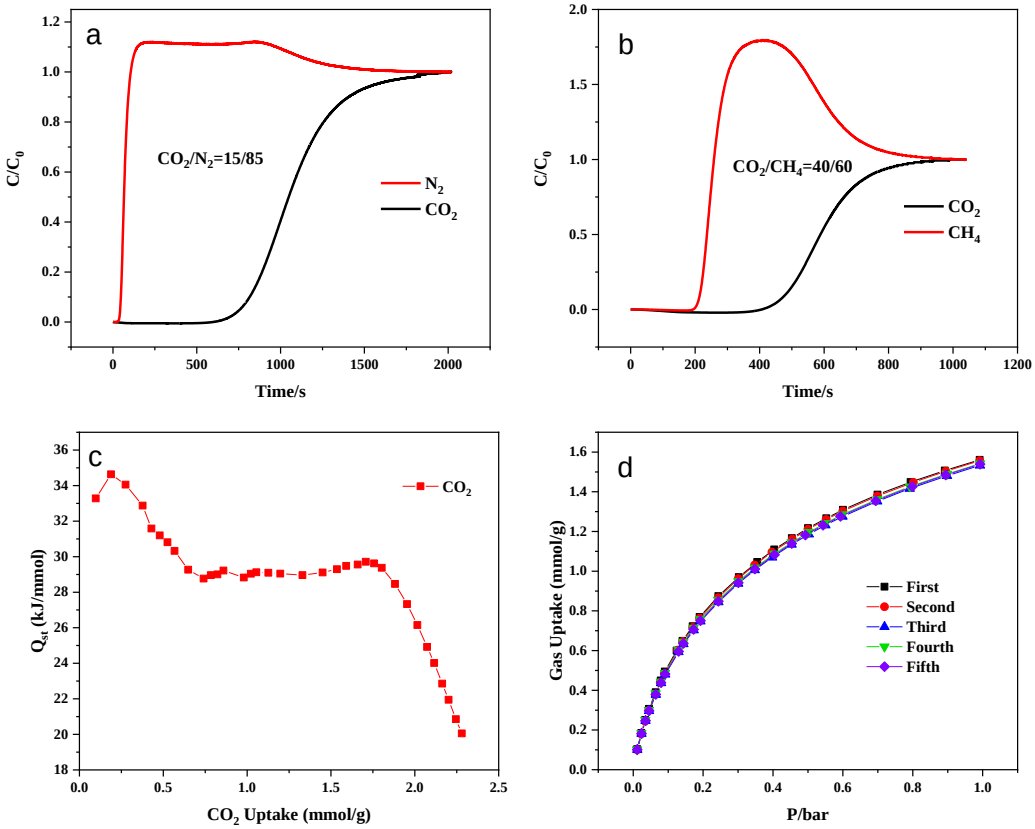

Fig. S12 Transient breakthrough curves for CO_2/N_2 (15/85) (a), CO_2/CH_4 (40/60) (b), isoteric heat of CO_2 adsorption Q_{st} (c) binary mixtures, regeneration test of N-CNs (d).