

## Electronic Supplementary Information

### Nanodiamond/carbon nitride hybrid nanoarchitecture as an efficient metal-free catalyst for oxidant- and steam-free dehydrogenation†

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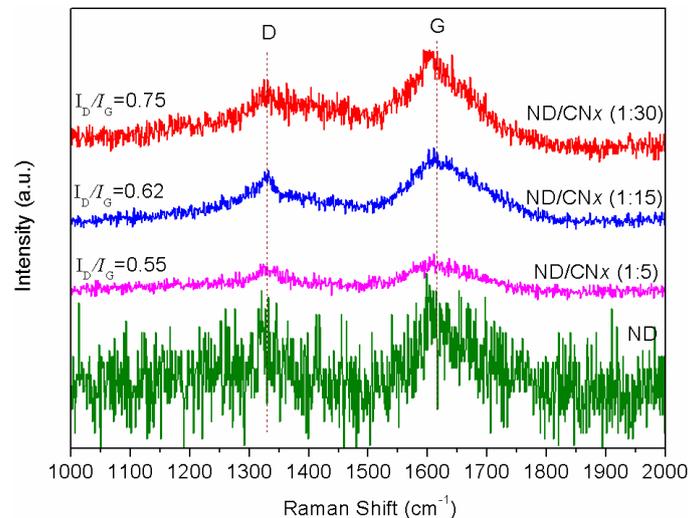
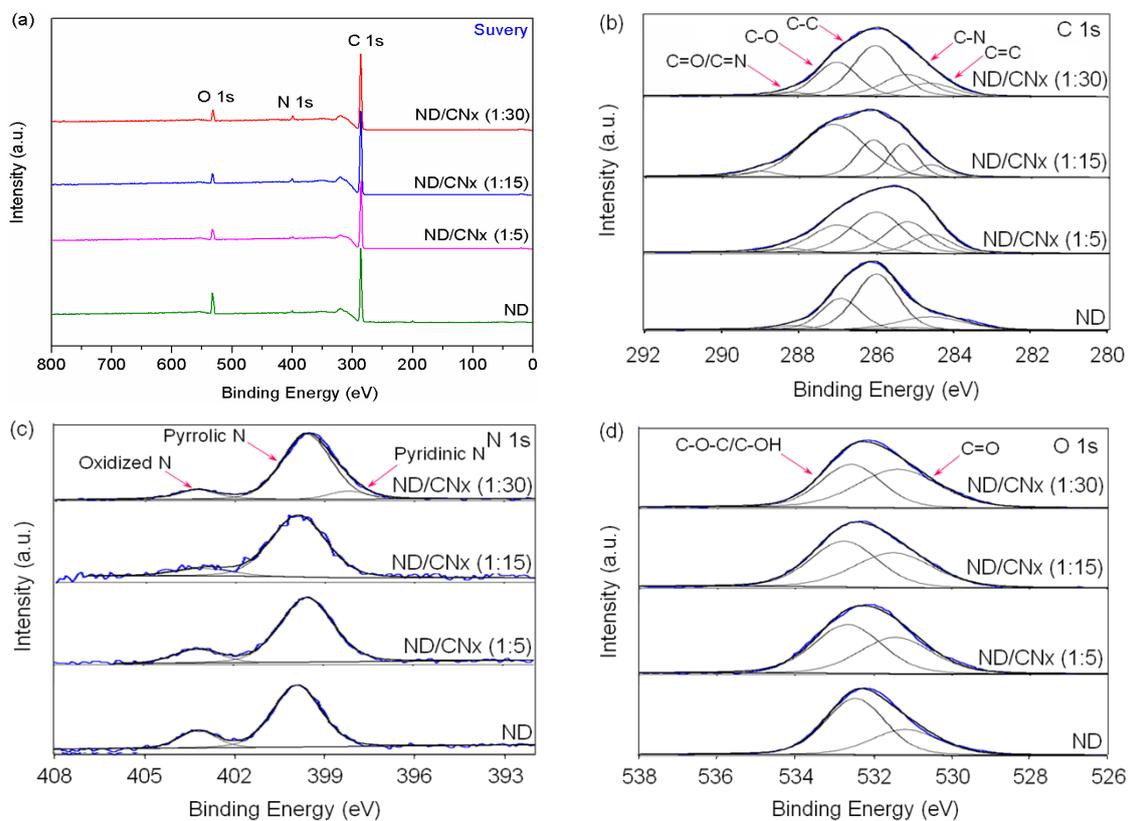


Fig. S1 Raman spectra of ND/CNx with diverse mass ratios of ND to melamine.



**Fig. S2** XPS spectra of ND/CNx nanostructures with diverse mass ratios of ND to melamines: (a) Survey spectra; (b) C 1s; (c) N 1s; (d) O 1s.

**Table S1.** Relative integrated intensity of the deconvoluted C 1s XPS spectra of ND/CNx with diverse pyrolysis temperatures, as well as the parent ND included for comparison.

Sample	C=C	C-N	C-C	C-O	C=O/C=N
	BE (eV)/Area <sup>a</sup> (%)				
ND	284.6/18.5	285.2/4.1	286.0/49.6	287.0/25.0	288.2/2.7
ND/CNx-650 <sup>b</sup>	284.6/26.6	285.2/2.0	286.0/15.1	-/-	288.1/53.7
ND/CNx-700	284.6/15.4	285.2/18.6	286.0/40.9	287.0/23.1	288.3/1.9
ND/CNx-750	284.6/11.9	285.2/19.3	286.0/41.4	287.0/25.5	288.3/1.8
ND/CNx-800	284.6/12.6	285.2/13.9	286.0/42.0	287.0/28.5	288.3/3.0

[a] Relative area percentage of various C species to the total C. [b] the peak appearing at 290 eV can be ascribed to absorbed CO<sub>2</sub> on the CNx layer (2.6% of percentage).

**Table S2.** Relative integrated intensity of the deconvoluted N 1s XPS spectra of ND/CNx with diverse pyrolysis temperatures, as well as the parent ND included for comparison.

Sample	Nitrogen	Pyridinic N	Pyrrolic N	Graphitic N	Oxidized N
	atom percentage <sup>a</sup> (%)	BE (eV)/Area <sup>b</sup> (%) / percentage <sup>c</sup> (%)	BE (eV)/Area <sup>b</sup> (%) / percentage <sup>c</sup> (%)	BE (eV)/Area <sup>b</sup> (%) / percentage <sup>c</sup> (%)	BE (eV)/Area <sup>b</sup> (%) / percentage <sup>c</sup> (%)
ND	1.5	-/-	399.9/82.2/1.2	-/-	403.2/17.8/0.3
ND/CNx-650 <sup>b</sup>	38.0	398.5/50.7/19.3	399.6/46.8/17.8	-/-	403.4/2.5/0.9
ND/CNx-700	5.4	398.2/23.0/1.2	399.6/74.1/4.0	-/-	403.2/2.9/0.2
ND/CNx-750	3.0	398.2/7.8/0.2	399.6/81.4/2.4	-/-	403.3/10.8/0.3
ND/CNx-800	2.5	-/-	399.6/85.2/2.1	-/-	403.1/14.8/0.4

[a] Nitrogen atom percentage in the sample. [b] Relative area percentage of various N species to the total N atoms. [c] Surface percentage in the samples of various N species.

**Table S3.** Relative integrated intensity of the deconvoluted O 1s XPS spectra of ND/CNx with diverse pyrolysis temperatures, as well as the parent ND included for comparison.

Sample	Oxygen	C=O	C-O-C/C-OH	C=O	C-O-C/C-OH
	atom percentage <sup>a</sup> (%)	BE (eV)/Area <sup>b</sup> (%)	BE (eV)/Area <sup>b</sup> (%)	percentage <sup>c</sup> (%)	percentage <sup>d</sup> (%)
ND	9.4	531.2/33.2	532.5/66.8	3.1	6.3
ND/CNx-650	3.1	531.4/48.0	532.6/52.0	1.5	1.6
ND/CNx-700	3.9	531.4/53.8	532.6/46.2	2.1	1.8
ND/CNx-750	5.1	531.4/54.6	532.6/45.4	2.7	2.3
ND/CNx-800	3.4	531.4/60.7	532.6/39.3	2.1	1.3

[a] Oxygen atom percentage in the sample. [b] Relative area percentage of various O species to the total O atoms. [c] Surface C=O percentage, calculated by total oxygen atom multiplies C=O relative area percentage to total O atoms. [d] Surface C-O-C/C-OH percentage, calculated by total oxygen atom multiplies C-O-C/C-OH relative area percentage to total O atoms.

**Table S4.** Relative integrated intensity of the deconvoluted C 1s XPS spectra of ND/CNx with diverse pyrolysis temperatures, as well as the parent ND included for comparison.

Sample	C=C	C-N	C-C	C-O	C=O/C=N
	BE (eV)/Area <sup>a</sup> (%)				
ND	284.6/18.5	285.2/4.1	286.0/49.6	287.0/25.0	288.2/2.7
ND/CNx (1:5)	284.6/12.9	285.2/23.1	286.0/36.2	287.0/24.6	288.3/3.3
ND/CNx (1:15)	284.6/15.4	285.3/18.6	286.1/40.9	287.1/23.1	289.0/1.9
ND/CNx (1:30)	284.6/11.9	285.2/19.3	286.0/41.4	287.0/25.5	288.3/1.8

[a] Relative area percentage of various C species to the total C.

**Table S5.** Relative integrated intensity of the deconvoluted N 1s XPS spectra of ND/CNx with diverse pyrolysis temperatures, as well as the parent ND included for comparison.

Sample	Nitrogen	Pyridinic N	Pyrrolic N	Graphitic N	Oxidized N
	atom percentage <sup>a</sup> (%)	BE (eV)/Area <sup>b</sup> (%) / percentage <sup>c</sup> (%)			
ND	1.5	-/-	399.9/82.2/1.2	-/-	403.2/17.8/0.3
ND/CNx (1:5)	5.1	-/-	399.6/84.7/4.3	-/-	403.2/15.3/0.9
ND/CNx (1:15)	3.8	-/-	399.9/88.6/3.4	-/-	403.1/11.4/0.4
ND/CNx (1:30)	3.0	398.2/7.8/0.2	399.6/81.4/2.4	-/-	403.3/10.8/0.3

[a] Nitrogen atom percentage in the sample. [b] Relative area percentage of various N species to the total N atoms. [c] Surface percentage in the samples of various N species.

**Table S6.** Relative integrated intensity of the deconvoluted O 1s XPS spectra of ND/CNx with diverse pyrolysis temperatures, as well as the parent ND included for comparison.

Sample	Oxygen	C=O	C-O-C/C-OH	C=O	C-O-C/C-OH
	atom percentage <sup>a</sup> (%)	BE (eV)/Area <sup>b</sup> (%)	BE (eV)/Area <sup>b</sup> (%)	percentage <sup>c</sup> (%)	percentage <sup>d</sup> (%)
ND	9.4	531.2/33.2	532.5/66.8	3.1	6.3
ND/CNx (1:5)	1.9	531.4/44.4	532.7/55.6	0.8	1.1
ND/CNx (1:15)	2.4	531.5/47.2	532.7/52.8	1.1	1.3
ND/CNx (1:30)	5.1	531.4/54.6	532.6/45.4	2.7	2.3

[a] Oxygen atom percentage in the sample. [b] Relative area percentage of various O species to the total O atoms. [c] Surface C=O percentage, calculated by total oxygen atom multiplies C=O relative area percentage to total O atoms. [d] Surface C-O-C/C-OH percentage, calculated by total oxygen atom multiplies C-O-C/C-OH relative area percentage to total O atoms.