

## Supplementary Material

### **Sustainable nonmetal catalyst based on sulfur-doped melamine-derived graphitic carbon nitride for CO<sub>2</sub> cycloaddition**

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**Table S1 Organic element analysis of CNS-X**

Entry	Catalyst	C (wt. %)	N (wt. %)	H (wt. %)	S (wt. %)
1	CNS-0.03	30.0	71.0	5.1	0.2
2	CNS-0.06	30.6	61.3	3.0	0.3
3	CNS-0.4	30.6	60.0	4.2	6.5
4	CNS-0.8	52.9	24.5	3.5	10.3
5	spent CNS-0.4	31.5	59.4	4.6	6.2

**Table S2 Independent variable factors and levels of simulation experiment.**

Factors	Symbols	Coded factors		
		Temperature-1	0	1
Temperature (°C)	A	110	125	140
Time (h)	B	2	6	10
CO <sub>2</sub> pressure (Mpa)	C	0.2	1.1	2
Catalyst amount (g)	D	0.05	0.2	0.35

**Table S3 Orthogonal experimental design based on RSM and corresponding results.**

Run	A: Temperature (°C)	B: Time (h)	C: CO <sub>2</sub> pressure (Mpa)	D: Catalyst amount (g)	R: Yield (%)
1	125	2	1.1	0.35	48.15
2	110	6	1.1	0.05	48.86
3	125	6	1.1	0.20	75.08
4	140	10	1.1	0.20	97.65
5	140	6	1.1	0.35	89.51
6	125	6	0.2	0.05	65.68
7	125	2	0.2	0.20	50.27
8	110	6	2.0	0.20	43.42
9	125	10	1.1	0.05	79.81
10	110	10	1.1	0.20	45.66
11	125	6	1.1	0.20	76.00
12	125	10	0.2	0.20	59.96
13	125	6	2.0	0.35	79.45
14	125	6	0.2	0.35	66.78
15	110	6	0.2	0.20	51.13
16	110	2	1.1	0.20	42.28
17	125	6	1.1	0.20	68.72

**Continued Table S3 Orthogonal experimental design based on RSM and corresponding results.**

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Run	A: Temperature (°C)	B: Time (h)	C: CO <sub>2</sub> pressure (Mpa)	D: Catalyst amount (g)	R: Yield (%)
18	140	6	1.1	0.05	83.27
19	125	6	1.1	0.20	69.98
20	140	2	1.1	0.20	57.13
21	125	6	2.0	0.05	53.27
22	125	10	1.1	0.35	91.90
23	140	6	0.2	0.20	68.76
24	125	10	2.0	0.20	87.64
25	125	6	1.1	0.20	74.20
26	125	2	2.0	0.20	37.59
27	140	6	2.0	0.20	78.02
28	110	6	1.1	0.35	48.03
29	125	2	1.1	0.05	40.76

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**Table S4 Quadratic model and ANOVA for yield (response surface) of Cyclic carbonate product.**

Model term	Sum of squares	Degrees of freedom	Mean square	F-value	P (P>F)
Model	7957.41	14	568.39	19.82	<0.0001
A-A	3167.49	1	3167.49	110.44	<0.0001
B-B	2897.35	1	2897.35	101.02	<0.0001
C-C	23.59	1	23.59	0.82	0.3798
D-D	226.79	1	226.79	7.91	0.0138
AB	344.91	1	344.91	12.03	0.0038
AC	71.89	1	71.89	2.51	0.1357
AD	12.48	1	12.48	0.44	0.5205
BC	407.18	1	407.18	14.20	0.0021
BD	5.53	1	5.53	0.19	0.6672
CD	157.29	1	157.29	5.48	0.0345
A <sup>2</sup>	182.73	1	182.73	6.37	0.0243
B <sup>2</sup>	333.60	1	333.60	11.63	0.0042
C <sup>2</sup>	297.87	1	297.87	10.39	0.0061
D <sup>2</sup>	0.052	1	0.052	0.0018	0.9665
Residual	401.54	14	28.68		
Lack of fit	359.49	10	35.95		
Pure error	42.05	4	10.51	3.42	0.1237
Total	8358.95	28			

**Table S5 Catalytic performance of CNS-0.4 for a series of CO<sub>2</sub> cycloaddition reactions**

Entry	Substrate	Time (h)	Conv. (%)	Select (%)
1	PO	4	72.1	98.8
2	ECH	4	76.9	98.4
3	SO	15	49.9	81.4
4	SO	30	77.8	85.7
5	CHO	30	9.6	76.5

Reaction conditions: epoxide 0.025 mol, acetonitrile 0.42 mol, toluene 0.009 mol, catalyst 0.2 g, 140 °C, 1 Mpa

**Table S6 Comparison of the catalytic performance for CO<sub>2</sub> cycloaddition over reported**

Entry	Catalyst	Time (h)	Temperature (°C)	Pressure (Mpa)	Yield (%)	Ref.
1	MCNB (0.01)	36	130	2.8	93.4	[1]
2	B <sub>0.1</sub> CN/SBA-15	24	130	3	94.7	[2]
3	M-41/CN	16	150	0.8	95.1	[3]
4	NUC-101/n-Bu <sub>4</sub> NBr	5	50	0.5	99.0	[4]
5	CNS-0.4	6	125	1	75.0	This work

## Conferences

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- [3] Liu D, Kang S, Xu Y, Duan P, Chen S. (2021). Core-shell catalyst with synergistic hydroxyl and nitrogen active sites for CO<sub>2</sub> cycloaddition, *J Environ. Chem. Eng.*, 9:106452-106460.
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