Supporting Information

Quick Drying Process: Promising Strategy for Preparing an Egg-shell-type Cu/γ-Al_2O_3 Catalyst for Direct N_2O Decomposition

Eun-Han Lee,[ab] In-Heon Kwak,[ac] Hansung Kim,[b] Shin-Kun Ryi,*[a]

[a] Korea Institute of Energy Research (KIER), 152 Gajeong-ro, Yuseong-gu, Daejeon 34129, Republic of Korea

[b] Department of Chemical and Biological Engineering, Yonsei University, 50 Yonsei-ro, Seodaemun-gu, Seoul 03722, Republic of Korea

[c] Department of Chemical and Biological Engineering, Korea University, 145 Anam-ro, Seongbuk-gu, Seoul, Republic of Korea

Table S1. Conditions of deN_2O tests.

	Balance	N_2O	NF_3	Steam	Temp. (^oC)	GHSV (h^{-1})
Case 1	N_2	1%	-	-	400-650	1,800
Case 2	Air		-	-	400-650	1,800
Case 3	Air		-	7.5%	400-650	1,960
Case 4	N_2	1-20%	-	-	450-600	1,800

Table S2. Cu shell thickness of the prepared catalysts.

Sample	Cu Shell thickness (mm)					
	1	2	3	4	5	Mean
OD-Cu(1)	0.99	1.05	1.00	0.97	0.99	1.00
VOD-Cu(1)	0.63	0.66	0.67	0.65	0.68	0.66
QD-Cu(1)	0.34	0.30	0.34	0.30	0.30	0.32
QD-Cu(5)	0.38	0.39	0.40	0.41	0.40	0.40
QD-Cu(10)	0.46	0.48	0.48	0.47	0.49	0.48
QD-Cu(15)	0.55	0.53	0.55	0.54	0.52	0.54

Table S3. Turnover frequency (TOF) value over OD-, VOD-, and QD-Cu(1)/γ-Al_2O_3 catalysts.

Catalyst	Temp.(℃)	r (μmols^{-1}g$_{cat}^{-1}$)	Active site (μmolg$_{cat}^{-1}$)	TOF (s^{-1})
OD-Cu(1)	450	0.011	135	7.8×10^{-5}
VOD-Cu(1)		0.014	139	9.9×10^{-5}
QD-Cu(1)		0.023	143	1.6×10^{-4}

Table S4. N_2O conversion over the QD-Cu(10)/γ-Al_2O_3 catalyst at 1% N_2O concentration and different feed conditions with time on stream.

Section	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Balance	N_2	Air	Air	Air	N_2	N_2	N_2	N_2	Air	Air	Air	N_2	N_2	N_2
Steam (%)	-	-	7.5	-	-	1.8 → 7.5	7.5	7.5	7.5	7.5	-	-	-	-
Temp. (°C)	500	500	500	500	500	550	550 →580	580	580	600	570	500	R.T	500
Conv. (%)	~99	~83	~54	~83	~99	~73	~98	~98	~96	~98	~99	~99	0	~99
Time (h)	24	24 -48	48 -72	72 -96	96 -161	161 -166	166 -171	171 -183	183 -190	190 -207	207 -231	231 -291	291 -313	313 -360

Table S5. N_2O conversion over the QD-Cu(10)/γ-Al_2O_3 catalyst at 20% N_2O concentration and different feed conditions with time on stream.

Section	**1**	**2**	**3**	**4**	**5**	**6**	**7**	8	9	10
Balance	N_2	Air	Air	Air	Air	Air	Air	N_2	N_2	N_2
Steam (%)	-	-	-	-	7.5	7.5	7.5	7.5	7.5	-
Temp. (°C)	550	550	550 →600	600	600	600 →630	630 →650	650	650 →640	640 →550
Conv. (%)	~99	~95	~99	~99	~96	~98	~99	~99	~99	~99
Time (h)	356	357 -358	358-372		373 -375	375 -376	377 -397	397 -399	400 -422	422 -500

Table S6. Long-term stability test of various catalysts in N_2O decomposition.

No.	Catalyst	Shape	Feed condition	GHSV (h^{-1})	T. (^oC)	Initial conv. value (%)	Durability (%)	Ref.
a	$Sm_{0.06}Ni$	Powder	2000 ppmv N_2O + 5 vol.% H_2O in Ar balance	20,000	350	59	56 after 8 h	[1]
b	$Ce_{20}CO$	Powder	1000 ppm N_2O in He balance	80,000	350	90	54 after 24 h	[2]
c	Co/MgO-15%	Powder	1% N_2O in He balance	30,000	500	100	100 after 100 h	[3]
d	$Pb_{0.04}Co$	Powder	2000 ppmv N_2O in Ar with or without 10 vol.% CO_2 + 5vol.% O_2	20,000	350	100	100 after 15 h	[4]
e	CuO	Powder	2600 ppm N_2O in He balance, p=0.3 MPa	19,000	480	100	5 after 15 h	[5]
	$Cu_{0.67}Ce_{0.33}O_y$	Powder	2600 ppm N_2O in He balance, p=0.3 MPa	19,000	480	100	91 after ~120 h	
f	Cu-ZSM-5	Powder	5000 ppm N_2O in He balance	-	475	80	80 after 50 h	[6]
g	γ-Al_2O_3	Pellets	1 mol.% N_2O in N_2 balance	1,818	700	100	100 after 350 h	[7]
h	Cu(10)/γ-Al_2O_3	Pellets	1 mol.% N_2O, 0-7.5% H_2O in N_2 or Air balance	1,800 -1,960	500-600	99	99 after 360 h	This study
i			20 mol.% N_2O, 0-7.5% H_2O in N_2 or Air balance	1,800 -1960	550-650	99	99 after 500 h	

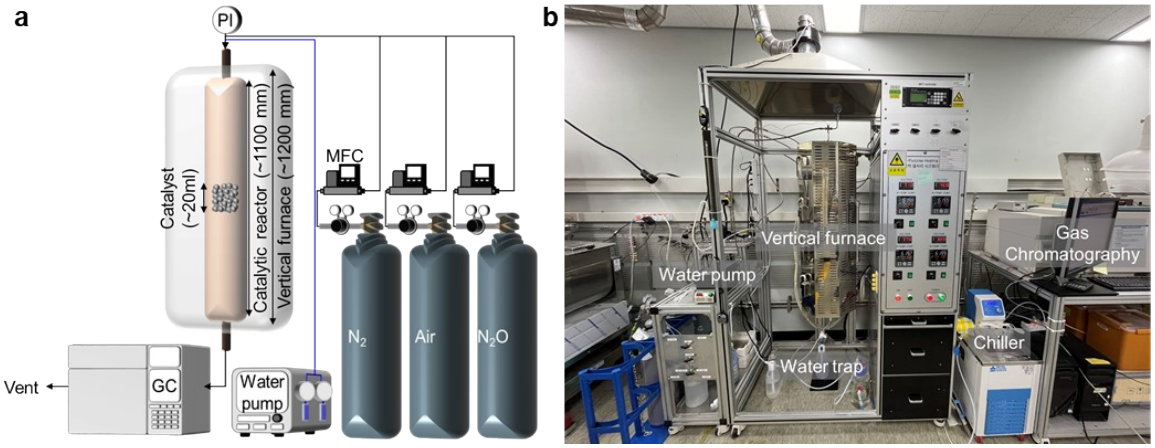


Fig. S1. (a) Schematic diagram and (b) optical image of the N_2O decomposition test system.

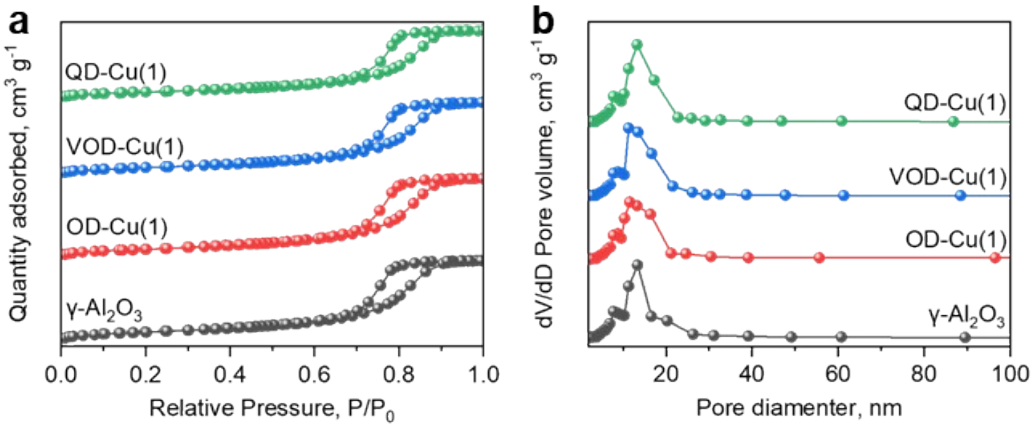


Fig. S2. (a) N_2 adsorption and desorption isotherms and (b) pore size distributions of the γ-Al_2O_3 and OD-, VOD-, and QD-Cu(1)/γ-Al_2O_3 catalysts.

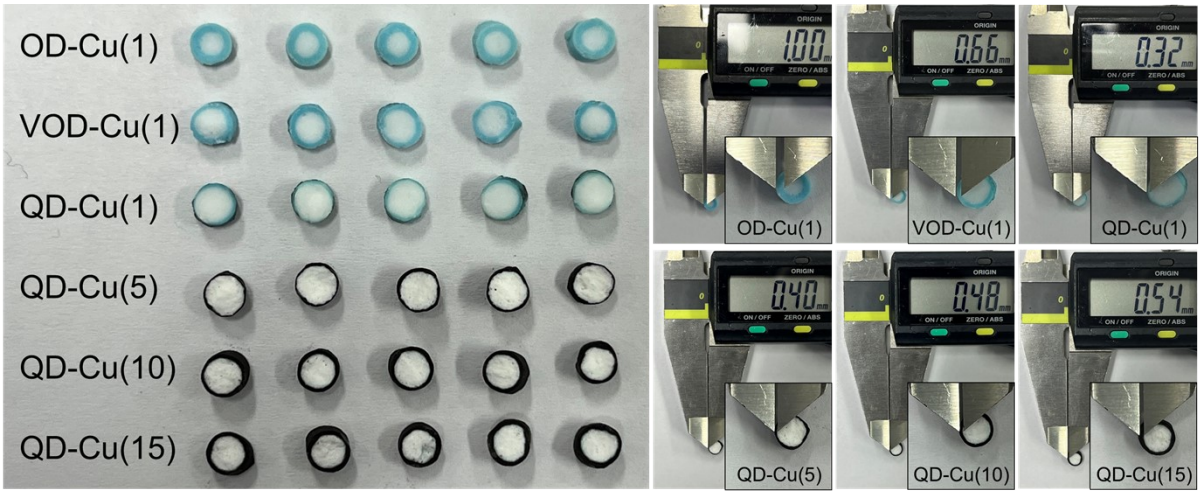


Fig. S3. Cross-sectional image of the prepared catalysts.

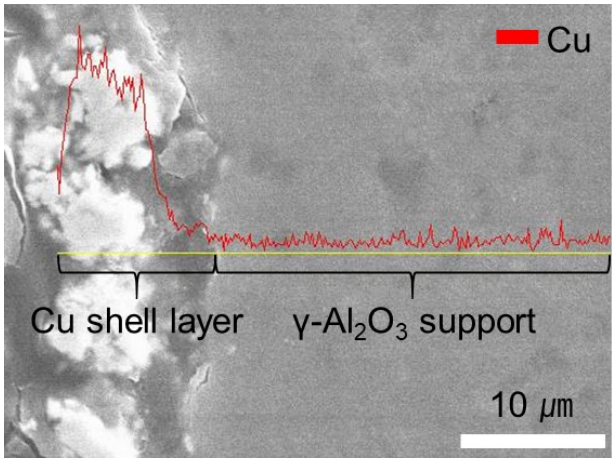


Fig. S4. EDS line scanning analysis of the QD-Cu(10)/γ-Al_2O_3 catalyst.

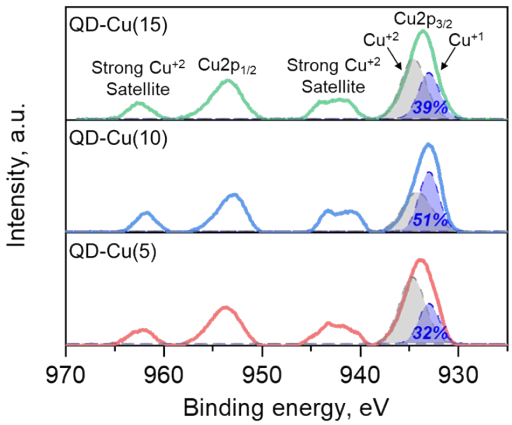


Fig. S5. XPS spectra of Cu 2p of the QD-Cu(x)/γ-Al_2O_3 catalysts (x: 5, 10, 15 wt.%).

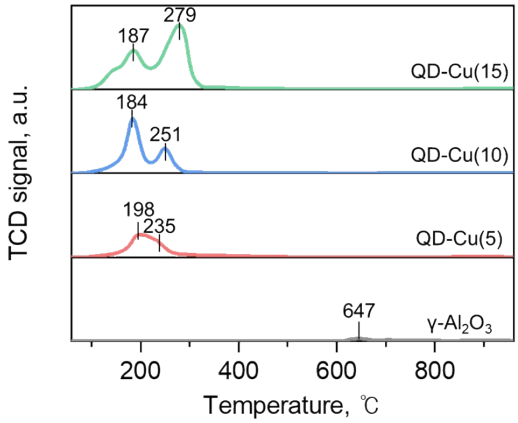


Fig. S6. H_2-TPR curves for the γ-Al_2O_3 and of the QD-Cu(x)/γ-Al_2O_3 catalysts (x: 5, 10, 15 wt.%).

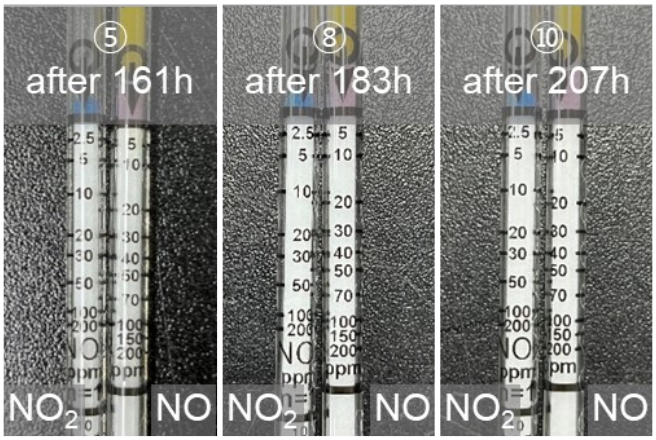


Fig. S7. NO_x (NO and NO_2) concentration at the outlet of the catalytic reactor measured by a gas detector during the deN_2O test using the QD-Cu(10)/γ-Al_2O_3 catalyst.

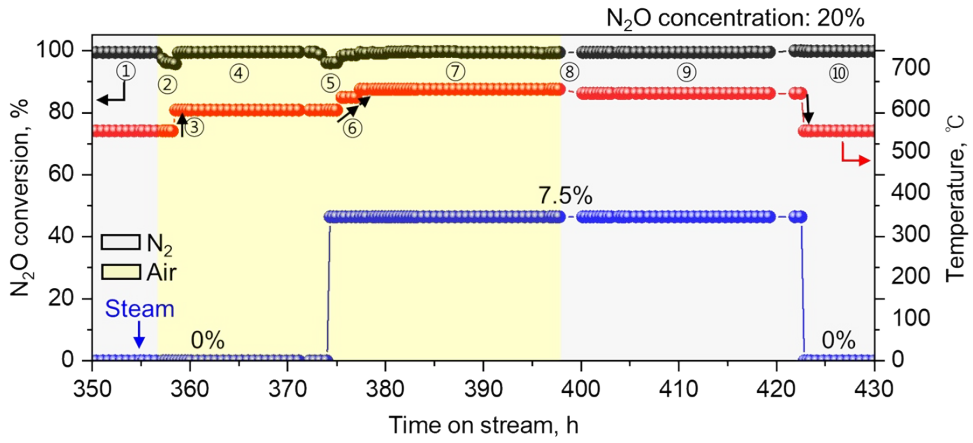
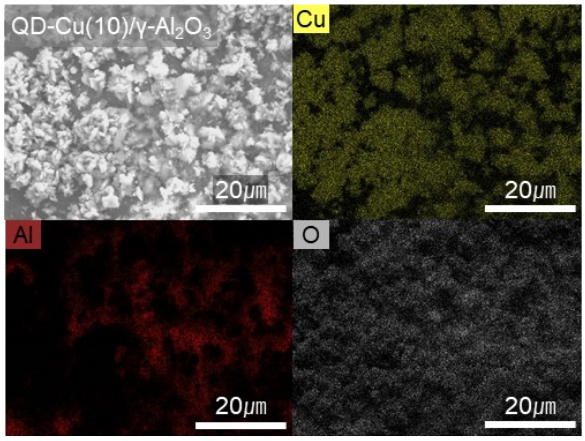


Fig. S8. N_2O conversion over the QD-Cu(10)/γ-Al_2O_3 catalyst at 20% N_2O concentration under different feed conditions with time on stream.



Element	Weight%
O K	32.88
Al K	16.46
Cu K	50.65
Totals	100.00

Fig. S9. SEM/EDS mapping analysis after 500hr stability test of the QD-Cu(10)/γ-Al_2O_3 catalyst surface.

References

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