

Supporting Information

Deactivation of bimetallic nickel-copper alloy catalyst in thermocatalytic decomposition of methane

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Table S1 Carbon yields and life span of the catalysts

Sample	Temperature (°C)	Average carbon deposition rate (mg _C /(g _{Ni} s).	Life span (h)	Theoretical carbon yield (g/g _{Ni})	Actual carbon yield (g/g _{Ni})
Ni/CNT	500	1.85	>64*	>424*	401*
	550	2.86	53	552	532
	575	3.85	27	377	358
	600	5.14	10	182	158
Ni ₈₇ Cu ₁₃ /CNT	600	2.72	>46*	>462*	451*
	650	4.33	36	579	562
	700	4.82	29	503	496
Ni ₇₈ Cu ₂₂ /CNT	600	3.48	>48*	>607*	601*
	650	4.46	48	768	718
	700	5.26	32	614	602
	750	5.54	13	271	230
Ni ₅₈ Cu ₄₂ /CNT	650	4.67	32	566	539
	700	4.99	29	529	506
	750	5.19	13	279	243
Ni ₄₇ Cu ₅₃ /CNT	650	4.32	32	538	498
	700	4.37	26	415	407
	750	4.72	12	234	206

Table S2 Structural properties of the resulting carbon nanofibers

Sample	Temperature (°C)	I _D /I _G (nm)	BET surface area (m ² /g)	Pore volume (cm ³ /g)
Ni/CNT	500	0.71	64	0.21
	550	0.74	70	0.21
	575	0.76	82	0.24
	600	0.78	86	0.25
Ni ₈₇ Cu ₁₃ /CNT	600	0.79	98	0.25
	650	0.83	110	0.27
	700	0.83	114	0.33
Ni ₇₈ Cu ₂₂ /CNT	600	0.81	107	0.32
	650	0.87	112	0.36
	700	0.92	134	0.36
	750	1.1	142	0.38
Ni ₅₈ Cu ₄₂ /CNT	650	0.94	127	0.37
	700	1.38	192	0.44
	750	1.47	246	0.46
Ni ₄₇ Cu ₅₃ /CNT	650	1.01	213	0.37
	700	1.52	237	0.47
	750	1.56	242	0.47

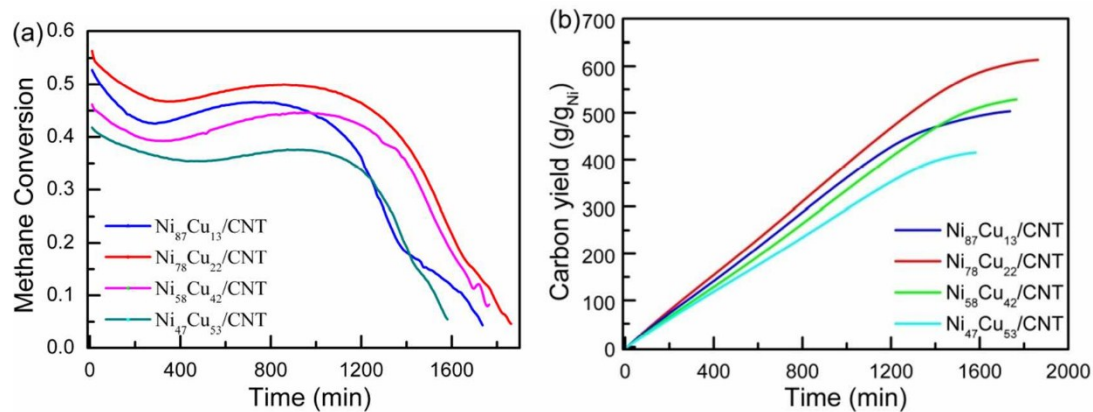


Figure S1 (a) Methane conversion and (b) carbon yield over the catalysts as a function of reaction time. Experimental conditions: 5 mg of metallic nickel was involved in the catalysts; pure methane with a volume flow rate of 7.5 ml min⁻¹; working temperature 700°C; weight hourly space velocity 59 g h⁻¹ g_{Ni}⁻¹.

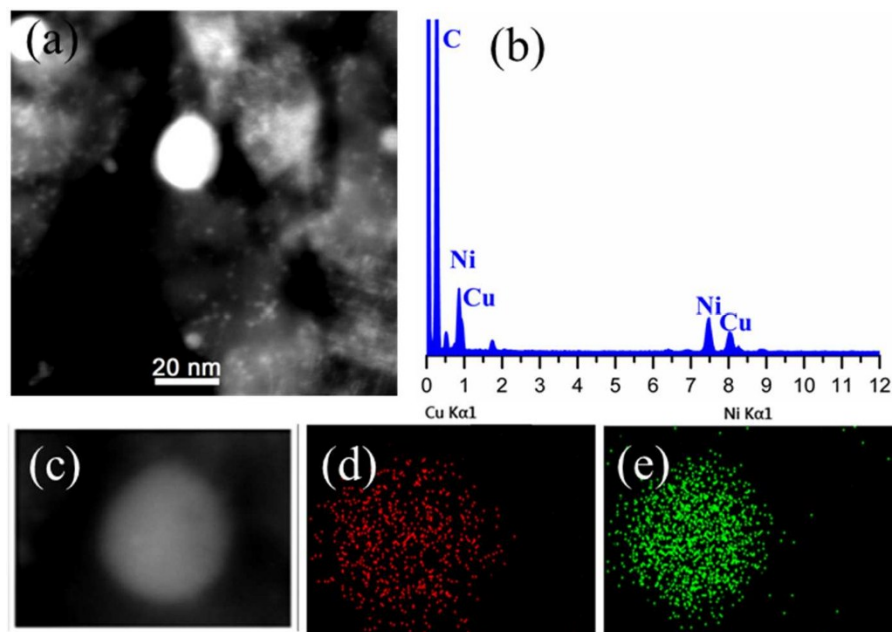


Figure S2 (a) HAADF-STEM micrograph of the spent Ni₇₈Cu₂₂/CNT catalyst at 650°C, (b) EDS profile of NiCu nanoparticle, (c) HAADF-STEM image of a NiCu nanoparticle, (d) and (e) corresponding EELS elemental mappings of Ni and Cu, respectively.

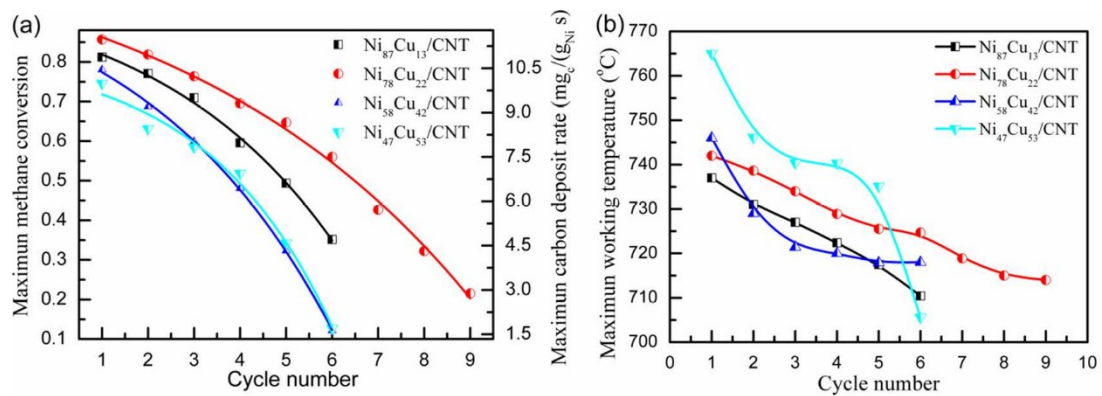


Figure S3 (a) Maximum methane conversion and (b) working temperature of the catalysts as a function of cycle number under a cyclic heating-cooling temperature mode.

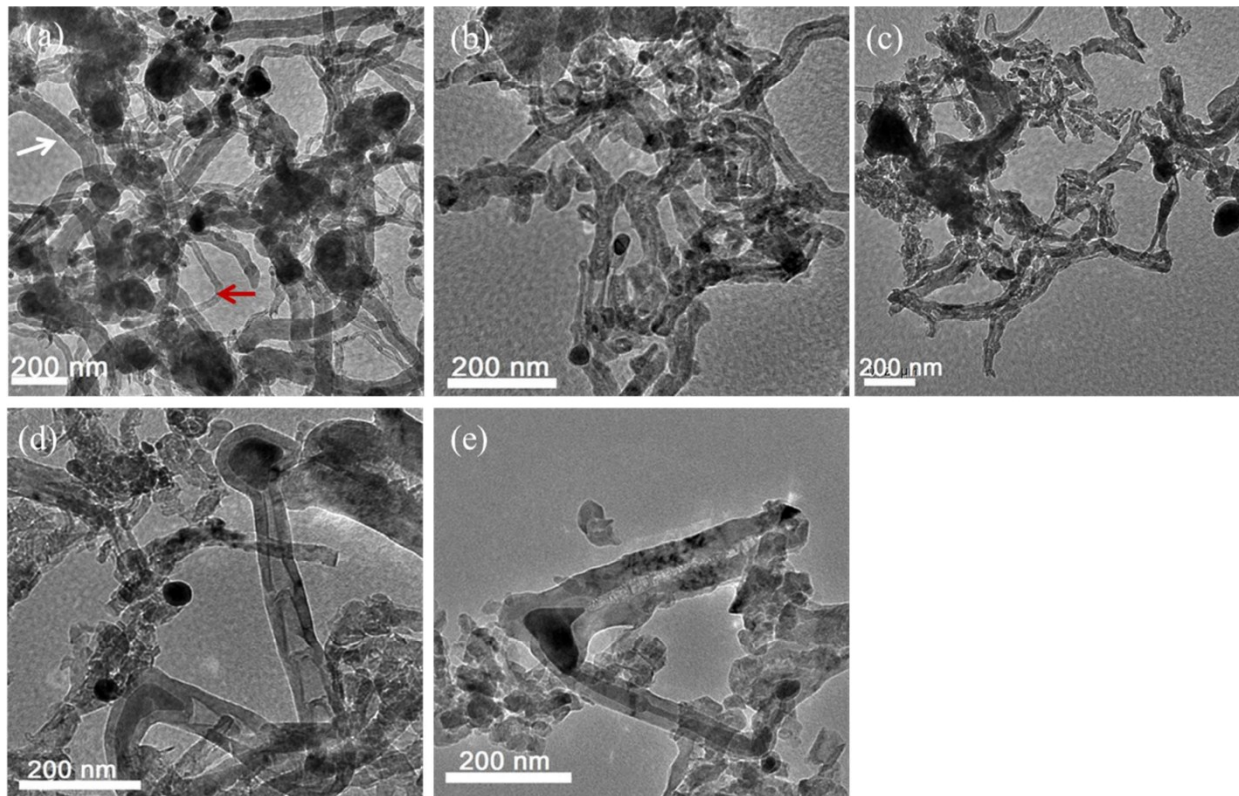


Figure S4 TEM images of deactivated (a) CNT, (b) $\text{Ni}_{87}\text{Cu}_{13}/\text{CNT}$, (c) $\text{Ni}_{78}\text{Cu}_{22}/\text{CNT}$, (d) $\text{Ni}_{58}\text{Cu}_{42}/\text{CNT}$ and (e) $\text{Ni}_{47}\text{Cu}_{53}/\text{CNT}$ under a cyclic heating-cooling temperature mode.