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Supplementary information

SiO_2@MnO_x@Na_2WO_4@SiO_2 core–shell-derived catalyst for oxidative coupling of methane

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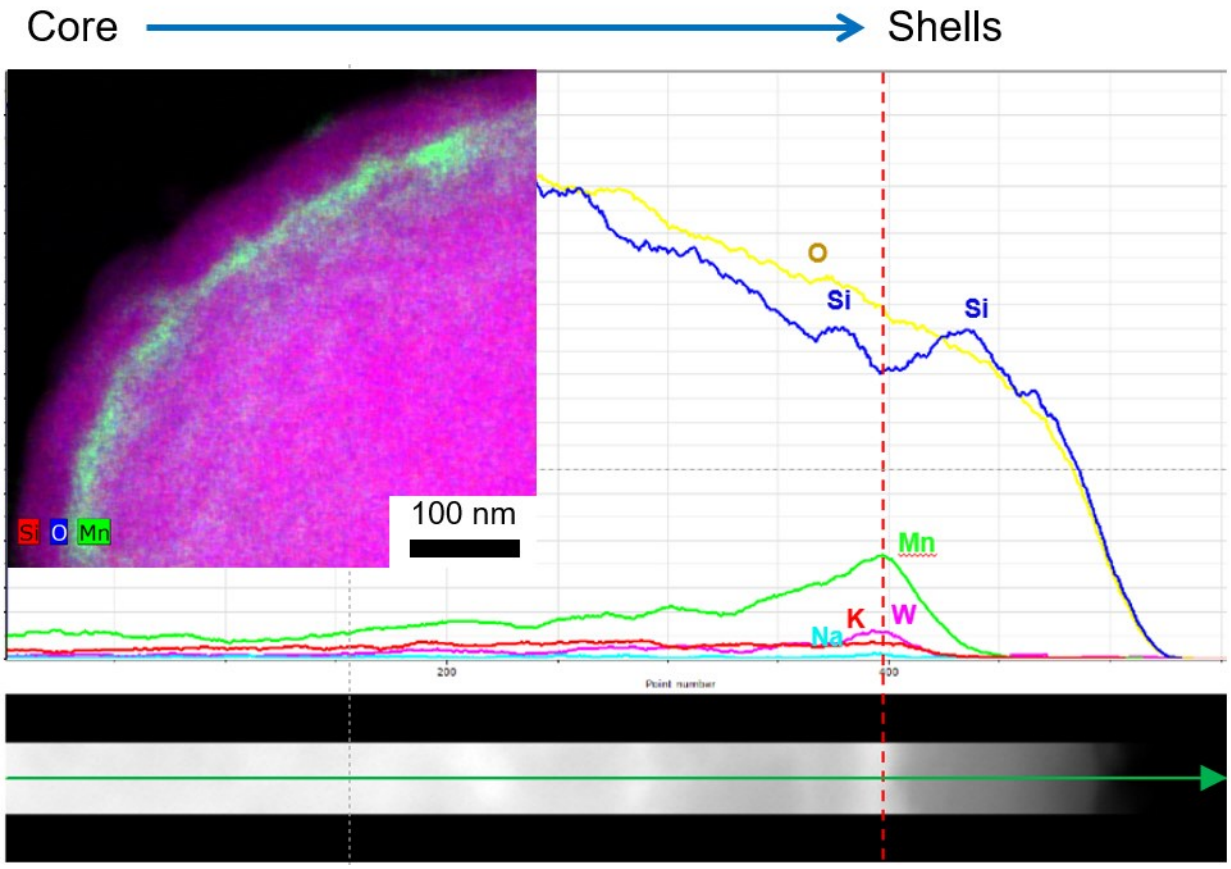

Fig. S1. EDS-STEM images of fresh SiO_2@Na_2WO_4@MnO_x($KMnO_4$)@SiO_2.

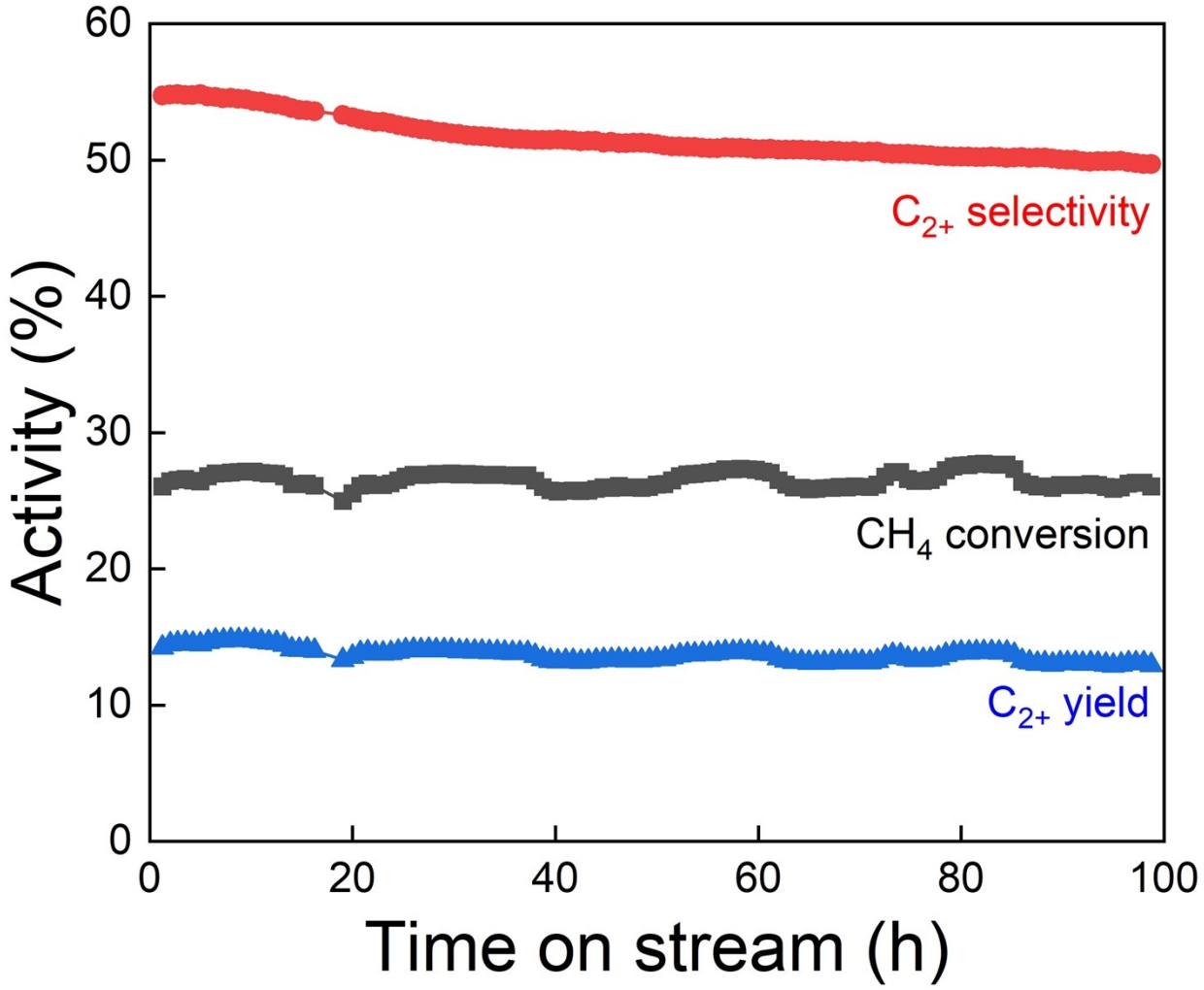

Fig. S2. Long term test results using SiO_2@MnO_x($KMnO_4$)@Na_2WO_4@SiO_2. (GHSV = 20,000 h^{-1}, temperature = 800 °C, $CH_4/O_2/N_2$ = 3/1/1 mol/mol/mol.)

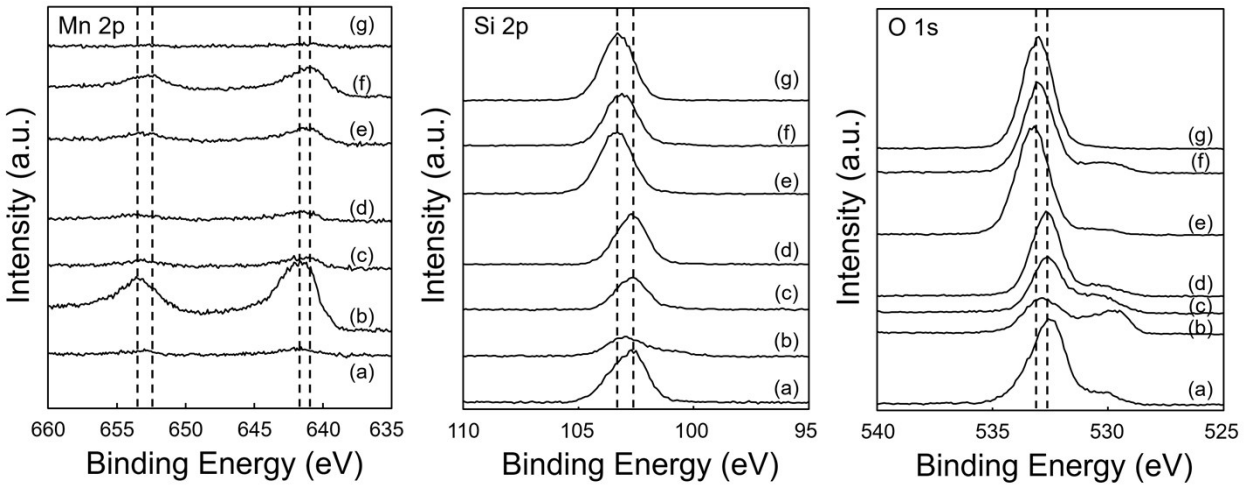

Fig. S3. Mn 2p, Si 2p, and O 1s XPS results of (a) Na_2WO_4/Mn/SiO_2, (b) SiO_2@MnO_x($KMnO_4$), (c) SiO_2@MnO_x($KMnO_4$)@Na_2WO_4, (d) SiO_2@MnO_x($KMnO_4$)@Na_2WO_4@SiO_2, (e) SiO_2@MnO_x(Mn-acetate), (f) SiO_2@MnO_x(Mn-acetate)@Na_2WO_4, and (g) SiO_2@MnO_x(Mn-acetate)@Na_2WO_4@SiO_2.

Table S1. OCM results depending on catalysts.[a]

Temperature (°C)	CH_4 conversion (%)	C_{2+} selectivity (%)	Olefins selectivity (%)	C_{2+} yield (%)	Olefins/paraffins (mol/mol)	Carbon balance (%)
$Na_2WO_4/Mn/SiO_2$						
740	23.3	52.3	34.3	12.2	1.9	98.5
770	29.1	52.8	37.7	15.3	2.5	101.4
800	30.3	48.2	38.5	14.6	4.0	94.7
$SiO_2@MnO_x(KMnO_4)@Na_2WO_4$						
740	4.3	49.8	17.9	2.2	0.6	116.5
770	9.5	49.7	27.0	4.7	1.2	103.5
800	17.0	46.4	35.2	7.9	3.1	103.6
$SiO_2@MnO_x(KMnO_4)@Na_2WO_4@SiO_2$						
740	10.8	54.7	26.7	5.9	1.0	109.6
770	19.5	53.8	37.2	10.5	2.2	107.0
800	25.4	50.8	40.3	12.9	3.8	105.3
$SiO_2@MnO_x$(Mn-acetate)$@Na_2WO_4@SiO_2$						
740	7.7	25.2	9.7	2.0	0.6	103.0
770	13.6	33.7	19.7	4.6	1.4	101.7
800	21.9	34.0	26.2	7.5	3.4	104.0

[a]GHSV = 20,000 h^{-1}, $CH_4/O_2/N_2$ = 3/1/1 mol/mol/mol.

Table S2. OCM results using SiO_2@MnO_x($KMnO_4$)@Na_2WO_4@SiO_2 depending on GHSV.[a]

Temperature (°C)	CH_4 conversion (%)	C_{2+} selectivity (%)	Olefins selectivity (%)	C_{2+} yield (%)	Olefins/paraffins (mol/mol)	Carbon balance (%)
GHSV = 20,000 h^{-1}						
740	10.8	54.7	26.7	5.93	1.0	109.6
770	19.5	53.8	37.2	10.5	2.2	107.0
800	25.4	50.8	40.3	12.9	3.8	105.3
GHSV = 30,000 h^{-1}						
740	7.1	50.3	18.5	3.56	0.6	100.6
770	14.2	53.1	30.7	7.5	1.4	98.2
800	23.5	55.5	40.2	13.1	2.6	103.8
GHSV = 50,000 h^{-1}						
740	3.3	52.1	11.9	1.73	0.6	110.4
770	7.4	58.2	22.7	4.3	1.4	101.9
800	15.0	60.5	36.9	9.1	3.4	101.6

[a]$CH_4/O_2/N_2$ = 3/1/1 mol/mol/mol.