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Supplementary Information

Direct Design of Active Catalysts for Low Temperature Oxidative Coupling of Methane via Machine Learning and Data Mining

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Table S1. The results of OCM at 500-900 °C using the mono metal oxides and Mn-Na_2WO_4/SiO_2 in Fig. 2(a).

Catalyst	Temp	CH_4 conv	O_2 conv	H_2 yield	CO yield	CO_2 yield	C_2H_4 yield	C_2H_6 yield	C_2 total
La_2O_3	400	1	1	0	0	0	0	0	0
	500	2	8	0	0	1	0	0	0
	600	26	89	2	6	9	6	7	13
	700	27	93	3	6	10	8	6	14
	800	29	98	4	6	11	11	4	15
	900	30	100	6	7	11	12	1	14
CeO_2	400	4	12	1	0	2	0	0	0
	500	17	26	1	0	16	0	0	0
	600	18	81	1	0	16	0	1	1
	700	18	95	2	1	16	0	1	1
	800	19	98	5	5	15	0	0	1
	900	24	100	11	14	12	1	0	1
Pr_6O_{11}	400	1	2	0	0	0	0	0	0
	500	18	99	0	0	16	1	2	3
	600	20	100	0	0	15	1	3	4
	700	21	100	0	0	15	2	4	6
	800	22	100	1	0	15	3	5	8
	900	24	100	2	2	14	7	2	8
Nd_2O_3	400	1	1	0	0	0	0	0	0
	500	27	95	3	4	11	6	7	14
	600	29	98	2	4	11	8	8	16
	700	30	100	3	4	12	9	8	17
	800	30	100	4	4	12	10	5	16
	900	27	100	5	5	12	9	1	10
Sm_2O_3	400	1	1	0	0	0	0	0	0
	500	3	11	1	0	2	0	0	0
	600	27	97	4	5	12	6	7	12
	700	29	99	4	5	12	7	7	14
	800	30	100	4	5	12	10	6	16
	900	29	100	6	6	12	12	2	14
Eu_2O_3	400	1	1	0	0	0	0	0	0
	500	28	98	3	5	12	6	7	13

	600	30	100	3	4	12	9	8	16
	700	30	100	3	4	12	9	8	17
	800	28	100	3	3	13	10	5	15
□	900	27	100	6	6	13	9	1	10
Gd_2O_3	400	1	0	0	0	0	0	0	0
	500	2	4	0	0	1	0	0	0
	600	16	58	5	6	7	1	3	5
	700	26	83	3	7	8	7	5	13
	800	29	96	4	7	10	11	4	15
□	900	28	99	5	7	11	11	1	12
Y_2O_3	400	1	0	0	0	0	0	0	0
	500	1	2	0	1	0	0	0	0
	600	23	79	5	9	8	3	5	8
	700	29	95	5	8	10	9	5	14
	800	29	98	6	8	11	10	3	13
□	900	29	100	8	9	11	10	1	11
Dy_2O_3	400	0	0	0	0	0	0	0	0
	500	0	4	1	0	0	0	0	0
	600	21	86	6	7	11	2	4	6
	700	28	97	4	7	11	8	6	14
	800	29	99	4	6	11	11	5	16
□	900	29	100	6	7	11	12	1	13
Yb_2O_3	400	-1	-1	0	0	0	0	0	0
	500	0	3	0	0	0	0	0	0
	600	9	84	4	7	3	0	1	2
	700	29	97	3	8	9	10	6	16
	800	29	99	4	8	10	11	5	16
□	900	28	100	7	11	11	10	2	11
MgO	400	1	1	0	0	0	0	0	0
	500	1	1	0	0	0	0	0	0
	600	3	10	2	2	1	0	0	0
	700	9	30	3	6	2	1	1	2
	800	16	48	3	9	2	4	3	7
□	900	28	91	7	16	6	9	2	11
Al_2O_3	400	1	1	0	0	0	0	0	0
	500	1	2	0	0	0	0	0	0

	600	1	11	0	0	0	0	0	0
	700	1	29	0	0	0	0	0	0
	800	5	43	1	2	0	1	2	3
□	900	26	91	7	20	2	8	2	10
ZnO	400	1	1	0	0	0	0	0	0
	500	2	6	1	0	1	0	0	0
	600	19	89	0	0	14	2	5	7
	700	21	95	0	0	14	3	5	8
	800	21	96	1	1	14	3	4	7
□	900	25	88	5	10	8	9	2	10
ZrO_2	400	0	0	0	0	0	0	0	0
	500	0	1	0	0	0	0	0	0
	600	3	12	2	4	1	0	0	0
	700	26	100	4	10	10	6	4	11
	800	26	100	5	10	10	7	3	10
□	900	27	100	6	10	11	8	1	9
Nb_2O_5	400	2	2	0	0	0	0	0	0
	500	2	1	0	0	0	0	0	0
	600	2	2	0	0	0	0	0	0
	700	2	3	0	0	0	0	0	0
	800	10	22	1	6	0	2	2	4
□	900	24	95	3	25	1	3	1	3
BaO	400	1	0	0	0	0	0	0	0
	500	0	2	0	0	0	0	0	0
	600	1	16	0	0	0	0	0	0
	700	1	95	0	0	0	0	0	0
	800	8	99	1	3	1	2	2	4
□	900	30	99	7	10	9	12	1	14
TiO_2	400	1	1	0	0	0	0	0	0
	500	1	2	0	0	0	0	0	0
	600	3	12	1	3	0	0	0	0
	700	22	94	4	21	4	0	0	0
	800	22	100	4	22	4	0	0	1
□	900	24	100	5	22	4	2	1	3
FeO	400	2	2	0	0	0	0	0	0
	500	2	2	0	0	0	0	0	0

	600	2	2	0	0	0	0	0	0
	700	3	11	0	0	1	0	0	0
	800	9	42	0	2	5	1	1	2
□	900	25	79	6	17	3	7	2	9
SnO	400	1	1	0	0	0	0	0	0
	500	1	3	0	0	0	0	0	0
	600	4	20	0	0	3	0	0	0
	700	17	96	0	0	15	0	2	2
	800	19	99	0	0	15	1	3	4
□	900	22	99	1	1	14	5	2	7
Mn-Na_2WO_4/SiO_2	400	1	1	0	0	0	0	0	0
	500	1	1	0	0	0	0	0	0
	600	1	1	0	0	0	0	0	0
	700	2	5	0	0	0	0	0	0
	800	23	69	3	11	3	9	3	12
	900	33	99	3	10	6	16	2	18

Table S2. The results of OCM at 500-900 °C using 1wt% M/La_2O_3 in Fig. 2(b). The loading of modifiers is 1wt%.

Catalyst	Temp	CH_4 conv	O_2 conv	H_2 yield	CO yield	CO_2 yield	C_2H_4 yield	C_2H_6 yield	C_2 total
Li/La_2O_3	400	1	0	0	0	0	0	0	0
	500	1	0	0	0	0	0	0	0
	600	1	2	0	0	0	0	0	0
	700	9	22	1	1	2	2	4	6
	800	29	99	3	3	12	10	5	16
	900	28	100	5	6	12	10	2	12
Na/La_2O_3	400	1	0	0	0	0	0	0	0
	500	1	2	0	0	0	0	0	0
	600	1	3	0	0	1	0	0	0
	700	29	98	3	3	12	9	7	17
	800	28	100	4	4	13	9	5	14
	900	26	100	6	7	12	8	1	9
Mg/La_2O_3	400	2	2	0	0	0	0	0	0
	500	22	91	0	3	11	3	5	8
	600	25	96	0	4	11	5	7	12
	700	28	98	2	3	12	8	7	15
	800	29	100	3	3	12	10	6	16
	900	28	100	5	4	12	11	2	13
K/La_2O_3	400	1	1	0	0	0	0	0	0
	500	1	1	0	0	0	0	0	0
	600	2	9	1	0	1	0	0	0
	700	30	100	3	3	13	9	8	17
	800	30	100	4	4	13	10	6	16
	900	27	100	5	5	13	10	2	12
Ca/La_2O_3	400	1	1	0	0	0	0	0	0
	500	30	97	3	4	11	8	8	17
	600	30	97	3	4	11	8	8	16
	700	31	98	3	4	11	9	7	17
	800	31	100	4	4	12	11	6	17
	900	30	100	6	5	12	12	1	14
V/La_2O_3	400	1	0	0	0	0	0	0	0
	500	1	3	0	0	0	0	0	0

	600	18	69	5	5	10	2	3	4
	700	24	93	5	6	13	4	4	8
	800	26	100	5	5	13	7	4	11
□	900	26	100	6	7	13	8	1	8
Cr/La_2O_3	400	1	1	0	0	0	0	0	0
	500	18	100	2	1	16	0	0	0
	600	19	100	2	2	16	0	1	2
	700	20	100	2	2	15	1	2	4
	800	22	100	2	2	15	3	3	6
□	900	23	100	4	3	14	5	1	6
Mn/La_2O_3	400	1	1	0	0	0	0	0	0
	500	1	7	0	0	1	0	0	0
	600	24	89	2	0	15	3	6	9
	700	25	93	2	1	15	4	6	11
	800	25	98	2	1	14	5	5	11
□	900	25	100	4	4	14	7	1	8
Fe/La_2O_3	400	1	2	0	0	0	0	0	0
	500	25	27	2	1	14	4	6	11
	600	26	33	1	1	14	6	8	13
	700	26	100	2	1	14	5	7	12
	800	25	100	3	2	14	6	5	11
□	900	24	100	5	5	13	6	1	8
Co/La2O3	400	1	2	0	0	0	0	0	0
	500	18	100	1	1	16	0	2	2
	600	19	100	1	1	16	1	2	3
	700	18	100	1	1	16	1	2	3
	800	19	100	1	1	15	1	2	3
□	900	20	100	4	4	14	2	1	3
Ni/La_2O_3	400	1	1	0	0	0	0	0	0
	500	27	100	16	9	18	0	2	2
	600	38	100	0	33	13	0	1	1
	700	50	100	0	57	6	0	0	0
	800	60	100	0	75	1	0	0	0
□	900	61	100	0	78	0	0	0	0
Cu/La_2O_3	400	1	2	0	0	0	0	0	0
	500	18	100	1	0	16	0	1	1

	600	18	100	1	0	16	0	1	1
	700	18	100	1	1	16	0	1	2
	800	20	100	3	2	15	1	2	4
□	900	24	100	8	8	14	3	1	4
Zn/La_2O_3	400	1	1	0	0	0	0	0	0
	500	29	96	4	4	12	7	8	15
	600	30	98	4	4	12	8	8	16
	700	30	98	3	4	12	9	7	17
	800	31	100	4	4	12	10	6	16
□	900	28	100	5	5	12	11	1	12
Rb/La_2O_3	400	1	0	0	0	0	0	0	0
	500	1	3	0	0	0	0	0	0
	600	20	68	3	3	8	5	5	10
	700	28	96	4	4	12	8	6	14
	800	30	100	5	4	13	11	5	16
□	900	30	100	8	7	12	13	1	14
Sr/La_2O_3	400	1	1	0	0	0	0	0	0
	500	30	97	3	3	12	9	9	17
	600	29	96	3	3	12	8	8	16
	700	30	98	3	3	12	10	8	17
	800	31	100	4	3	12	12	6	18
□	900	30	100	6	5	12	13	1	15
Mo/La_2O_3	400	1	1	0	0	0	0	0	0
	500	23	89	2	5	11	4	5	9
	600	23	98	2	5	12	3	5	8
	700	24	100	2	5	13	4	5	9
	800	27	100	3	5	12	7	5	13
□	900	27	100	6	6	12	10	1	11
Pd/La_2O_3	400	18	100	2	4	15	0	0	0
	500	20	100	6	7	14	0	0	0
	600	23	100	10	11	14	0	0	0
	700	28	100	17	18	13	0	0	0
	800	28	100	16	20	12	0	0	0
□	900	37	100	0	37	8	0	0	1
Ag/La_2O_3	400	1	2	0	0	0	0	0	0
	500	23	29	0	0	15	3	6	9

	600	26	72	1	1	14	6	7	14
	700	28	96	2	2	13	8	8	15
	800	28	100	3	3	13	9	6	15
□	900	27	100	5	5	13	10	2	12
In/La_2O_3	400	1	1	0	0	0	0	0	0
	500	25	87	1	3	11	6	7	14
	600	27	99	1	1	13	8	8	15
	700	27	99	1	1	13	8	7	15
	800	28	100	1	1	13	9	6	15
□	900	27	100	4	2	13	11	2	13
Sn/La_2O_3	400	1	0	0	0	0	0	0	0
	500	1	86	0	0	0	0	0	0
	600	9	97	3	4	4	0	1	2
	700	23	100	1	3	11	4	5	9
	800	27	100	1	3	12	9	5	14
□	900	28	100	4	5	12	12	1	14
Cs/La_2O_3	400	1	1	0	0	0	0	0	0
	500	2	5	1	0	1	0	0	0
	600	20	74	4	4	10	3	4	7
	700	29	98	4	4	13	8	7	14
	800	30	100	5	4	13	11	5	16
□	900	30	100	8	7	12	13	1	14
Ba/La_2O_3	400	1	1	0	0	0	0	0	0
	500	20	17	3	2	9	4	6	10
	600	27	28	2	3	12	7	8	15
	700	29	39	3	3	12	9	8	17
	800	31	99	4	3	13	12	6	18
□	900	29	100	7	6	13	12	2	13
W/La_2O_3	400	1	1	0	0	0	0	0	0
	500	20	72	3	5	8	4	5	9
	600	26	91	4	6	11	6	6	12
	700	27	95	5	5	12	6	6	12
	800	30	99	5	5	12	11	5	16
□	900	30	100	7	7	12	13	1	14
Re/La_2O_3	400	1	1	0	0	0	0	0	0
	500	18	100	2	3	15	0	1	2

	600	18	100	2	2	15	0	1	2
	700	20	100	2	2	15	1	3	4
	800	24	99	3	3	14	3	3	6
□	900	25	100	6	7	13	7	1	8
Pt/La_2O_3	400	1	1	0	0	0	0	0	0
	500	1	2	0	0	0	0	0	0
	600	19	100	2	2	15	0	2	2
	700	21	100	3	4	14	0	4	4
	800	25	100	8	9	14	1	3	4
□	900	28	100	14	17	12	3	1	3

Table S3. The results of OCM at 500-900 °C using 1 wt% In modified rare earth oxides, 0.5-0.01wt% In/La_2O_3, 0.1wt% In/Nd_2O_3, and 0.1wt% In/Eu_2O_3 in Fig. 2(c).

Catalyst	Temp	CH_4 conv	O_2 conv	H_2 yield	CO yield	CO_2 yield	C_2H_4 yield	C_2H_6 yield	C_2 total
In/CeO_2	400	16	100	0	0	16	0	0	0
	500	17	98	0	0	16	0	0	0
	600	17	99	1	0	16	0	0	0
	700	18	100	3	2	16	0	1	1
	800	20	100	6	6	15	0	0	1
□	900	26	100	13	17	12	1	0	1
In/Pr_6O_{11}	400	1	3	0	0	0	0	0	0
	500	19	100	0	0	16	1	3	3
	600	18	100	0	0	16	1	2	3
	700	19	100	0	0	16	1	3	4
	800	20	100	0	0	15	2	3	5
□	900	22	100	2	1	15	5	1	6
In/Nd_2O_3	400	1	1	0	0	0	0	0	0
	500	27	99	1	1	13	6	8	14
	600	25	99	2	2	14	5	7	11
	700	27	100	1	1	13	7	8	14
	800	28	100	2	1	13	9	6	15
□	900	26	100	3	2	13	9	2	11
In/Eu_2O_3	400	1	1	0	0	0	0	0	0
	500	26	98	1	3	13	5	7	12
	600	27	97	1	2	13	7	8	14
	700	27	100	1	1	14	7	7	15
	800	26	100	1	1	14	8	5	13
□	900	25	100	4	4	13	8	1	9
In/Sm_2O_3	400	1	1	0	0	0	0	0	0
	500	27	98	1	2	13	6	8	14
	600	26	97	1	2	12	6	7	13
	700	27	100	1	2	13	7	8	14
	800	27	100	1	1	13	9	6	15
□	900	27	100	4	3	13	11	2	12
In/Gd_2O_3	400	1	1	0	0	0	0	0	0
	500	20	83	1	3	11	4	5	8

	600	25	96	1	2	12	6	7	12
	700	25	100	1	1	13	7	7	14
	800	26	100	1	1	13	7	6	13
□	900	24	100	2	2	14	8	2	10
In/Dy_2O_3	400	0	0	0	0	0	0	0	0
	500	2	8	0	0	1	0	0	0
	600	22	87	1	2	11	4	6	10
	700	24	98	0	1	14	5	7	11
	800	25	100	1	1	14	6	6	12
□	900	24	100	2	2	13	8	2	10
In/Y_2O_3	400	1	1	0	0	0	0	0	0
	500	19	83	1	3	11	2	4	6
	600	24	96	1	2	13	5	6	11
	700	25	100	1	1	13	6	7	13
	800	26	100	1	1	13	8	6	14
□	900	26	100	3	3	13	9	1	11
In/Yb_2O_3	400	-1	0	0	0	0	0	0	0
	500	0	1	0	0	0	0	0	0
	600	14	57	0	2	7	2	3	6
	700	21	87	0	1	12	5	5	10
	800	22	96	1	1	13	5	5	10
□	900	22	99	2	2	14	7	2	8
0.5wt% In/La_2O_3	400	1	2	0	0	0	0	0	0
	500	26	89	1	2	11	6	7	14
	600	27	95	1	2	12	7	8	15
	700	27	100	1	1	13	8	7	15
	800	27	100	2	1	13	8	6	14
□	900	26	100	4	3	13	9	2	11
0.1wt% In/La_2O_3	400	1	1	0	0	0	0	0	0
	500	29	96	3	4	11	8	8	17
	600	30	98	3	4	12	9	8	18
	700	30	99	3	3	12	10	8	18
	800	30	100	3	3	12	10	6	17
□	900	28	100	5	4	13	11	2	13

0.01wt% In/La_2O_3	400	2	1	0	0	0	0	0	0
	500	3	8	1	1	1	0	0	0
	600	28	92	3	4	11	8	8	15
	700	29	98	3	4	12	9	7	16
	800	29	100	4	4	13	10	6	16
□	900	28	100	6	5	13	10	2	12
0.1wt% In/Nd_2O_3	400	1	1	0	0	0	0	0	0
	500	27	98	2	3	12	6	7	13
	600	28	99	2	3	13	6	7	14
	700	29	100	2	2	13	8	8	15
	800	29	100	2	2	13	10	6	16
□	900	28	100	4	3	13	12	2	13
0.1wt% In/Eu_2O_3	400	1	1	0	0	0	0	0	0
	500	27	99	3	4	12	6	7	12
	600	27	99	4	4	13	5	7	12
	700	29	100	3	4	12	7	7	14
	800	29	100	3	3	12	10	6	16
□	900	29	100	7	7	12	12	1	13

Table S4. The results of OCM at 500-900 °C using the predicted catalysts in Fig. 5. The loading of modifiers is 1wt%.

Catalyst	Temp	CH_4 conv	O_2 conv	H_2 yield	CO yield	CO_2 yield	C_2H_4 yield	C_2H_6 yield	C_2 total
Al/Y_2O_3	400	1	1	0	0	0	0	0	0
	500	18	74	6	7	10	1	2	3
	600	22	82	7	9	10	2	3	5
	700	24	87	6	10	9	4	4	8
	800	28	96	5	7	11	9	4	13
	900	28	100	6	8	11	10	2	12
Ca/Y_2O_3	400	1	0	0	0	0	0	0	0
	500	1	2	0	0	0	0	0	0
	600	4	60	2	2	1	0	0	0
	700	31	93	4	5	11	11	7	18
	800	31	98	4	5	12	12	6	18
	900	30	100	6	6	12	13	2	15
Ni/Y_2O_3	400	1	1	0	0	0	0	0	0
	500	35	99	0	27	14	0	0	0
	600	44	100	0	45	9	0	0	0
	700	54	100	0	65	4	0	0	0
	800	60	100	0	75	1	0	0	0
	900	61	100	0	77	0	0	0	0
Zn/Y_2O_3	400	15	89	0	0	14	0	0	0
	500	16	98	0	0	16	0	0	0
	600	17	100	0	0	16	0	0	0
	700	17	100	0	0	16	0	1	1
	800	18	100	0	0	16	0	2	2
	900	1	1	0	0	0	0	0	0
Ag/Y_2O_3	400	1	3	0	0	2	0	0	0
	500	16	31	0	0	15	0	2	2
	600	21	89	0	0	15	2	4	6
	700	24	96	1	0	14	5	6	11
	800	27	98	2	2	13	8	5	14
	900	29	100	5	6	12	11	2	13

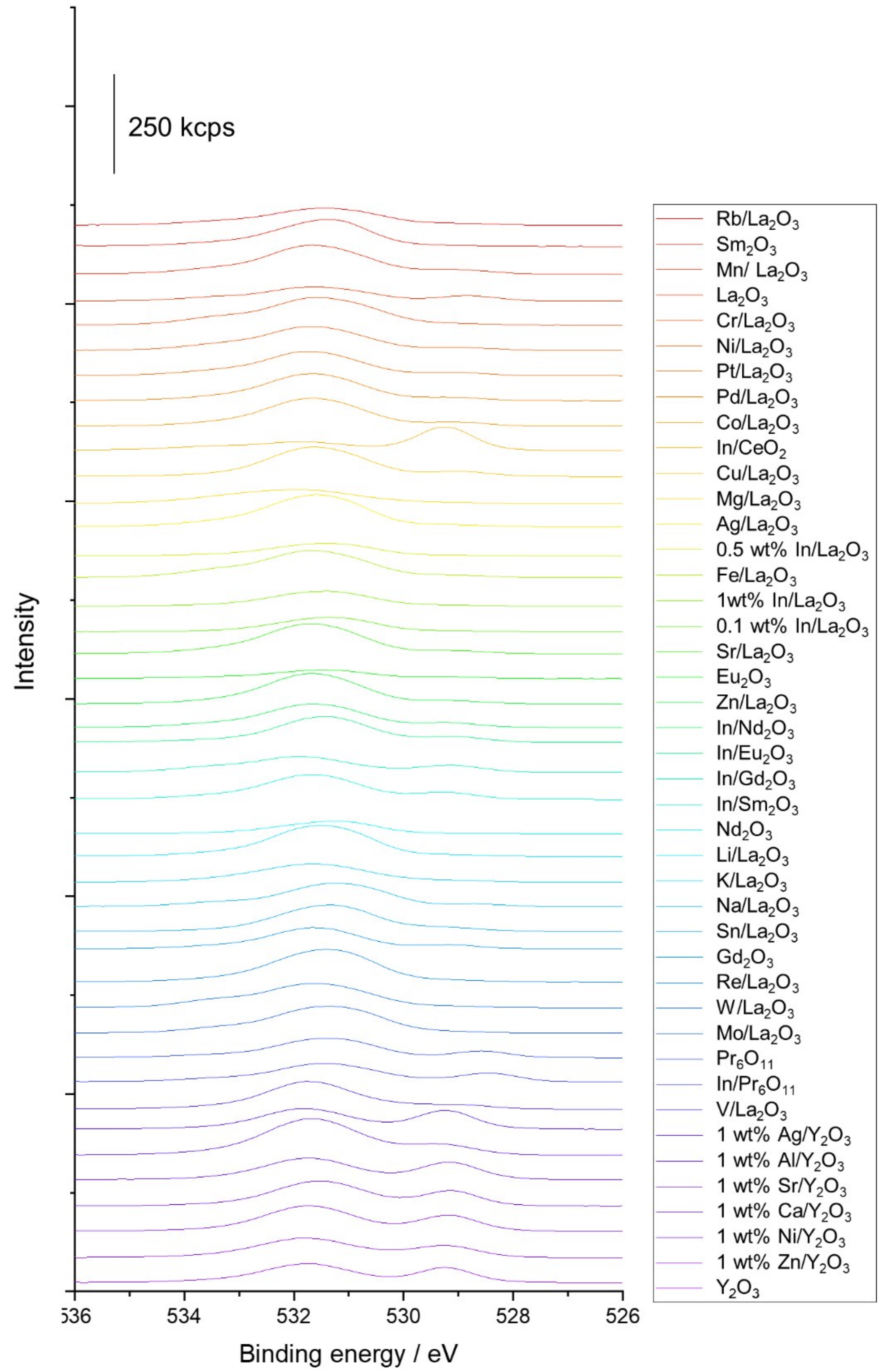

Figure S1. O 1s XPS of the catalysts analyzed.

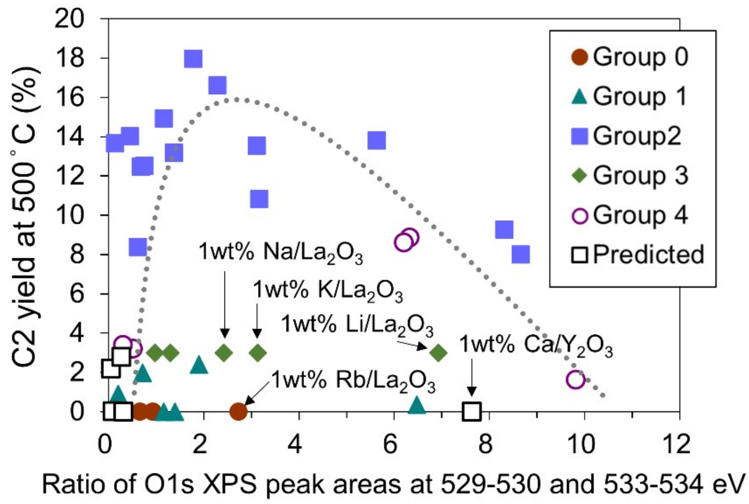

Figure S2. Dependence of the C2 yield at 500 °C on the ratio of O1s XPS peak areas at 529-530 and 533-534 eV of the catalysts. Group 0: ●, Group 1: ▲, Group 2: ■, Group 3: ♦, Group 4: ○, Predicted: □.

Table S5. Compounds used in this work.

Compound	Supplier
$LiNO_3$	FUJIFILM Wako Pure Chemical
$NaNO_3$	FUJIFILM Wako Pure Chemical
$Mg(NO_3)_2 \cdot 6H_2O$	FUJIFILM Wako Pure Chemical
KNO_3	FUJIFILM Wako Pure Chemical
$Ca(NO_3)_2 \cdot 4H_2O$	FUJIFILM Wako Pure Chemical
NH_4VO_3	FUJIFILM Wako Pure Chemical
$Cr(NO_3)_2 \cdot 9H_2O$,	Sigma-Aldrich
$Mn(NO_3)_2 \cdot 6H_2O$	FUJIFILM Wako Pure Chemical
$Fe(NO_3)_3 \cdot 9H_2O$	FUJIFILM Wako Pure Chemical
$Co(NO_3)_2 \cdot 6H_2O$	FUJIFILM Wako Pure Chemical
$Ni(NO_3)_2 \cdot 6H_2O$	FUJIFILM Wako Pure Chemical
$Cu(NO_3)_2 \cdot 2H_2O$	FUJIFILM Wako Pure Chemical
$Zn(NO_3)_2 \cdot 6H_2O$	Kishida Chemical
$RbNO_3$	FUJIFILM Wako Pure Chemical
$Sr(NO_3)_2 \cdot 6H_2O$	FUJIFILM Wako Pure Chemical
$(NH_4)_6Mo_7O_{24} \cdot 4H_2O$	FUJIFILM Wako Pure Chemical
$Pd(NO_3)_2$	FUJIFILM Wako Pure Chemical
$AgNO_3$	FUJIFILM Wako Pure Chemical
$In(NO_3)_3 \cdot 3H_2O$	FUJIFILM Wako Pure Chemical
$SnCl_2 \cdot 2H_2O$	NACALAI TESQUE
$CsNO_3$	FUJIFILM Wako Pure Chemical
$Ba(NO_3)_2$	FUJIFILM Wako Pure Chemical
$(NH_4)_{10}W_{12}O_{41} \cdot 5H_2O$	Kishida Chemical
NH_4ReO_4	FUJIFILM Wako Pure Chemical
$Pt(NO_3)_2 \cdot (NH_3)_2$	Tanaka Kikinzoku Kogyo
Y_2O_3	Kishida Chemical
La_2O_3	Kishida Chemical
CeO_2	DAIICHI KIGENSO KAGAKU KOGYO
Pr_6O_{11}	FUJIFILM Wako Pure Chemical
Nd_2O_3	Kishida Chemical
Sm_2O_3	Kishida Chemical
Eu_2O_3	FUJIFILM Wako Pure Chemical
Gd_2O_3	Kishida Chemical
Dy_2O_3	FUJIFILM Wako Pure Chemical

Yb_2O_3	FUJIFILM Wako Pure Chemical
αAl_2O_3	FUJIFILM Wako Pure Chemical
BaO	FUJIFILM Wako Pure Chemical
ZnO	FUJIFILM Wako Pure Chemical
ZrO_2	Kojundo Chemical Lab.
Nb_2O_5	FUJIFILM Wako Pure Chemical
TiO2（JRC-TIO-6）	Catalysis Society of Japan
FeO	FUJIFILM Wako Pure Chemical
SnO	FUJIFILM Wako Pure Chemical
SiO_2 (60N)	Kanto Chemical
$Na_2WO_4 \cdot 2H_2O$	Sigma-Aldrich