Electronic Supplementary Information

Reaction condition optimization for non-oxidative conversion of methane using artificial intelligence

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Table S1. Experimental data of MTOAH used in this study.

Index	Pressure (barg)	Temperature (°C)	Flow rate (sccm)	H_2 content	Reactor length (cm)	Reactor diameter (mm)	CH_4 conversion (%)	C_2 selectivity (%)	Coke selectivity (%)	C_2 yield (%)
1	0.031	890.1	40	0	45	16	2.07	41.01	12.79	0.85
2	0.069	920	80	0	45	16	1.89	46.81	16.06	0.89
3	0.024	890	120	0	45	16	2.55	37.84	35.13	0.97
4	0.114	939.9	120	0	45	16	2.56	44.39	26.57	1.14
5	0.032	900.2	40	0	45	16	3.57	38.28	15.61	1.37
6	0.05	919.9	60	0	45	16	3.69	38.93	31.05	1.44
7	0.07	930.1	80	0	45	16	3.96	39.48	25.16	1.56
8	0.024	899.9	120	0	45	16	4.42	39.08	28.01	1.73
9	0.116	950	120	0	45	16	5.16	39.48	25.36	2.04
10	0.032	910.1	40	0	45	16	6.02	35.36	21.56	2.13
11	0.367	1120	320	1	15	6.8	2.72	78.36	10.52	2.13
12	0.05	929.8	60	0	45	16	6.57	35.13	30.79	2.31
13	0.07	940.2	80	0	45	16	7.1	34.64	30.1	2.46
14	0.367	1125	320	1	15	6.8	3.24	76.33	14.52	2.47
15	0.024	909.8	120	0	45	16	7.67	33.65	35.46	2.58
16	0.033	920.2	40	0	45	16	9.28	31.12	30.37	2.89
17	0.05	939.9	60	0	45	16	10.42	30.71	35.3	3.2
18	0.368	1135	320	1	15	6.8	3.78	86	6.67	3.25
19	0.025	919.9	120	0	45	16	11.92	27.49	43.74	3.28
20	0.831	1125	80	1	15	16	34.2	10.01	77.24	3.42
21	0.866	1120	80	1	15	16	32.06	10.72	75.56	3.44
22	0.795	1130	80	1	15	16	35.66	9.68	78.1	3.45
23	0.755	1135	80	1	15	16	37.94	9.15	79.5	3.47
24	0.885	1115	80	1	15	16	30.24	11.61	73.66	3.51
25	0.033	930.1	40	0	45	16	13.1	26.94	39.19	3.53
26	0.708	1140	80	1	15	16	39.5	9.04	80.19	3.57
27	0.664	1145	80	1	15	16	41.38	8.74	81.05	3.62
28	0.873	1110	80	1	15	16	28.79	12.57	71.84	3.62
29	0.025	929.9	120	0	45	16	15.92	23.07	49.65	3.67
30	0.009	1150	80	1	15	16	40.79	9.34	78.66	3.81
31	0.025	939.9	120	0	45	16	19.67	19.51	55.26	3.84
32	0.026	949.9	120	0	45	16	23.09	16.64	60.46	3.84
33	0.027	950	120	0	45	16	23.39	16.43	60.59	3.84
34	0.005	1155	80	1	15	16	41.18	9.34	79.3	3.85
35	0.052	949.8	60	0	45	16	14.81	26.12	41.88	3.87
36	0.034	940.1	40	0	45	16	17.22	22.6	48.38	3.89
37	0.111	1105	80	2	45	4	5.5	77.23	20.68	4.25
38	0.467	1115	160	1	15	6.8	5	87.76	6.1	4.39
39	0.12	1134.9	80	1	15	4	5.14	92.01	3.81	4.73
40	0.119	1105.1	80	1	15	6.8	5.14	93.25	0.81	4.79
41	0.193	1170	240	5	15	16	5	96.09	2.09	4.8
42	0.37	1150	320	1	15	6.8	5.81	83	9.14	4.83
43	0.286	1145.1	160	3	15	6.8	5.87	83.32	16.68	4.89
44	0.111	1110	80	2	45	4	6.22	79.29	18.1	4.93
45	0.56	1075	240	1	15	16	5.44	91.27	0.88	4.96
46	0.468	1120	160	1	15	6.8	5.68	88.11	5.9	5
47	0.273	1098	240	0.5	15	16	5.6	93.35	3.79	5.23
48	0.158	1125	80	3	15	6.8	6.89	76.91	11	5.3
49	0.367	1155	320	1	15	6.8	6.5	83.4	10.12	5.42
50	0.119	1140	80	1	15	4	5.99	90.65	3.96	5.43
51	0.286	1150.1	160	3	15	6.8	6.18	88.49	10.77	5.47
52	0.12	1114.9	80	1	30	4	6.13	90.18	4.54	5.53
53	0.256	1140	120	2	15	6.8	6.38	88.06	5.33	5.62
54	0.111	1115	80	2	45	4	6.9	82.46	14.83	5.69
55	0.188	1100	240	3	15	16	6.16	94.02	2.02	5.79
56	0.072	1065	80	1	15	16	8.02	72.99	13.28	5.85
57	0.274	1103	240	0.5	15	16	6.22	94.21	2.09	5.86
58	0.193	1175	240	5	15	16	5.95	98.57	0.36	5.87
59	0.157	1130	80	3	15	6.8	6.98	84.24	3.07	5.88
60	0.286	1155	160	3	15	6.8	6.84	88.38	10.7	6.04
61	0.868	1080	240	1	15	16	7.82	77.77	7.14	6.08
62	0.37	1160	320	1	15	6.8	7.24	85.26	8.72	6.17

Table S1. (Continued)

Index	Pressure (barg)	Temperature (°C)	Flow rate (sccm)	H_2 content	Reactor length (cm)	Reactor diameter (mm)	CH_4 conversion (%)	C_2 selectivity (%)	Coke selectivity (%)	C_2 yield (%)
63	0.121	1144.9	80	1	15	4	6.79	91.06	1.13	6.18
64	0.189	1105	240	3	15	16	6.79	92.01	3.2	6.24
65	0.119	1120	80	1	30	4	7.36	84.88	8.81	6.25
66	0.194	1177.5	240	5	15	16	6.5	97.84	0.86	6.36
67	0.306	1105	80	1	15	16	27.1	23.64	56.6	6.4
68	0.156	1135	80	3	15	6.8	7.87	81.73	7.38	6.43
69	0.11	1120	80	2	45	4	7.86	81.92	15.27	6.44
70	0.272	1109	240	0.5	15	16	7.25	89.13	6.31	6.46
71	0.469	1130	160	1	15	6.8	7.13	90.84	0.46	6.47
72	0.75	1085	240	1	15	16	8.72	75.3	8.2	6.57
73	0.286	1160.1	160	3	15	6.8	7.23	92.78	6.03	6.71
74	0.072	1070	80	1	15	16	10.2	65.97	16.29	6.73
75	0.615	1180	240	3	15	16	25.71	26.29	59.94	6.76
76	0.189	1110	240	3	15	16	7.25	93.49	0.53	6.78
77	0.193	1180	240	5	15	16	7.25	94.9	3.52	6.88
78	0.257	1150	120	2	15	6.8	7.89	88.2	5.8	6.96
79	0.121	1149.9	80	1	15	4	8.02	87.49	5.86	7.01
80	0.566	1090	240	1	15	16	9.05	77.55	5.69	7.02
81	0.374	1165	320	1	15	6.8	7.93	88.53	3.49	7.02
82	0.548	1185	240	3	15	16	28.22	25.05	62.33	7.07
83	0.888	1115	240	1	15	16	19.32	36.72	39.13	7.1
84	0.209	1100	80	1	15	16	24.74	28.76	49.85	7.12
85	0.12	1124.9	80	1	30	4	8.42	84.57	7.29	7.12
86	0.272	1113	240	0.5	15	16	8.05	88.89	5.35	7.15
87	0.157	1140	80	3	15	6.8	7.8	91.77	1	7.16
88	0.531	1190	240	3	15	16	30.57	23.46	65.16	7.17
89	0.522	1195	240	3	15	16	32.37	22.37	67.26	7.24
90	0.11	1125	80	2	45	4	8.7	83.48	13.64	7.26
91	0.47	1135	160	1	15	6.8	8.37	86.92	3.13	7.28
92	0.12	1120	80	1	15	6.8	8.7	83.92	4.7	7.3
93	0.188	1115	240	3	15	16	8	91.25	1.58	7.3
94	0.075	1075	80	1	15	16	11.78	62.09	16.87	7.31
95	0.286	1165.1	160	3	15	6.8	8.1	91.37	7.2	7.4
96	0.478	1095	240	1	15	16	10.16	73.32	9.71	7.45
97	0.193	1182.5	240	5	15	16	7.88	95.77	2.38	7.54
98	0.078	1080	80	1	15	16	14.4	53.02	24.74	7.63
99	0.189	1120	240	3	15	16	8.74	88.31	3.02	7.72
100	0.129	1095	80	1	15	16	21.44	36.3	40.44	7.78
101	0.156	1145	80	3	15	6.8	9.6	81.72	8.87	7.84
102	0.373	1170	320	1	15	6.8	9.23	85.25	6.3	7.87
103	0.272	1119	240	0.5	15	16	8.71	90.39	1.55	7.87
104	0.088	1085	80	1	15	16	16.38	48.17	27.93	7.89
105	0.105	1090	80	1	15	16	18.75	42.39	33.54	7.95
106	0.122	1154.9	80	1	15	4	9.34	85.11	6.13	7.95
107	0.441	1100	240	1	15	16	11.24	71.52	9.62	8.04
108	0.119	1130	80	1	30	4	9.75	83	7.23	8.1
109	0.193	1185	240	5	15	16	8.68	93.6	4.29	8.12
110	0.47	1140	160	1	15	6.8	9.51	85.7	2.28	8.15
111	0.111	1129.9	80	2	45	4	9.64	84.72	10.94	8.17
112	0.287	1170.1	160	3	15	6.8	8.72	93.95	4.23	8.2
113	0.19	1125	240	3	15	16	9.6	86.37	3.33	8.29
114	0.119	1124.9	80	1	15	6.8	10.08	82.98	1.93	8.36
115	0.257	1160	120	2	15	6.8	9.65	87.06	7.55	8.4
116	0.387	1105	240	1	15	16	12.27	69.41	10.49	8.52
117	0.385	1160	240	1	15	6.8	10.02	85.1	5.51	8.52
118	0.272	1123	240	0.5	15	16	9.89	86.58	3.68	8.56
119	0.157	1150	80	3	15	6.8	9.71	88.66	0.24	8.61
120	0.193	1187.5	240	5	15	16	8.96	96.43	0.96	8.64
121	0.55	1110	240	1	15	16	15.24	56.79	19.44	8.66
122	0.189	1130	240	3	15	16	10.76	81.16	7.21	8.73
123	0.374	1175	320	1	15	6.8	10.56	83.4	6.93	8.81
124	0.122	1159.9	80	1	15	4	10.56	84.39	5.06	8.91
125	0.139	1180	180	5	15	16	10.08	89.28	6.42	9
126	0.138	1180	180	5	15	16	10.49	86.15	9.52	9.03

Table S1. (Continued)

Index	Pressure (barg)	Temperature (ºC)	Flow rate (sccm)	H_2 content	Reactor length (cm)	Reactor diameter (mm)	CH_4 conversion (%)	C_2 selectivity (%)	Coke selectivity (%)	C_2 yield (%)
127	0.12	1134.9	80	1	30	4	11.33	79.81	8.47	9.04
128	0.286	1175.1	160	3	15	6.8	9.27	97.57	0.1	9.05
129	0.47	1145	160	1	15	6.8	11.04	82.05	4.3	9.06
130	0.139	1180	180	5	15	16	10.28	88.22	7.22	9.07
131	0.139	1180	180	5	15	16	10.22	88.84	6.75	9.08
132	0.193	1190	240	5	15	16	10.23	89.01	8.04	9.11
133	0.111	1135	80	2	45	4	10.68	86.07	8.79	9.19
134	0.257	1165	120	2	15	6.8	10.42	88.29	5.87	9.2
135	0.188	1135	240	3	15	16	11.72	78.61	7.76	9.21
136	0.12	1130	80	1	15	6.8	11.93	77.86	6.55	9.29
137	0.273	1128	240	0.5	15	16	10.99	84.51	3.4	9.29
138	0.177	1180	240	5	15	16	10.28	90.67	5.42	9.32
139	0.158	1180	210	5	15	16	10.26	91.1	4.49	9.35
140	0.123	1180	150	5	15	16	11.16	84.03	10.06	9.38
141	0.158	1180	210	5	15	16	10.38	90.49	5.01	9.39
142	0.161	1180	210	5	15	16	10.4	90.46	5.06	9.4
143	0.178	1180	240	5	15	16	10.29	91.51	4.51	9.41
144	0.177	1180	240	5	15	16	10.28	91.68	4.29	9.43
145	0.177	1180	240	5	15	16	10.51	89.93	6.1	9.45
146	0.156	1155	80	3	15	6.8	11.25	84.01	5.08	9.45
147	0.386	1165	240	1	15	6.8	11.53	83.24	5.23	9.6
148	0.188	1140	240	3	15	16	12.83	75.32	9.21	9.66
149	0.123	1180	150	5	15	16	11.34	85.53	8	9.7
150	0.123	1180	150	5	15	16	11.19	87.02	6.08	9.74
151	0.193	1192.5	240	5	15	16	10.68	91.89	4.51	9.81
152	0.375	1180	320	1	15	6.8	11.63	84.61	3.51	9.84
153	0.122	1180	150	5	15	16	11.55	85.31	7.73	9.86
154	0.287	1180.1	160	3	15	6.8	11.06	89.52	7.5	9.9
155	0.472	1150	160	1	15	6.8	12.99	76.3	8.68	9.91
156	0.273	1133	240	0.5	15	16	12.12	81.97	3.58	9.94
157	0.122	1165	80	1	15	4	12.41	80.17	6.21	9.95
158	0.081	1180	60	5	15	16	14.78	67.81	20.81	10.02
159	0.187	1145	240	3	15	16	13.98	71.81	11.38	10.04
160	0.108	1180	120	5	15	16	12.42	80.9	10.47	10.05
161	0.257	1170	120	2	15	6.8	11.62	86.48	6.27	10.05
162	0.081	1180	60	5	15	16	14.84	67.82	20.64	10.06
163	0.081	1180	60	5	15	16	14.93	67.42	21.3	10.06
164	0.121	1139.9	80	1	30	4	13.27	75.87	10.45	10.07
165	0.108	1180	120	5	15	16	12.27	82.21	8.94	10.09
166	0.108	1180	120	5	15	16	12.34	82.07	9.22	10.12
167	0.111	1140	80	2	45	4	12.31	82.37	11.44	10.14
168	0.081	1180	60	5	15	16	15.1	67.2	21.81	10.15
169	0.094	1180	90	5	15	16	13.72	74.17	15.01	10.17
170	0.094	1180	90	5	15	16	13.64	74.64	14.51	10.18
171	0.094	1180	90	5	15	16	14	72.99	16.36	10.22
172	0.156	1160	80	3	15	6.8	11.91	86.15	3.6	10.26
173	0.188	1150	240	3	15	16	15.39	66.91	14.89	10.29
174	0.094	1180	90	5	15	16	13.62	75.87	13.17	10.33
175	0.119	1134.9	80	1	15	6.8	13.57	76.33	3.81	10.36
176	0.193	1195	240	5	15	16	12.06	86.26	9.79	10.4
177	0.274	1137	240	0.5	15	16	13.85	75.74	8.08	10.49
178	0.387	1170	240	1	15	6.8	13.34	79.03	7.32	10.55
179	0.19	1155	240	3	15	16	17.02	62.15	19.07	10.58
180	0.191	1160	240	3	15	16	18.79	56.69	24.47	10.65
181	0.205	1170	240	3	15	16	22.33	48.19	33.8	10.76
182	0.473	1155	160	1	15	6.8	15.04	71.6	11.66	10.77
183	0.194	1165	240	3	15	16	20.42	52.76	28.56	10.77
184	0.288	1185.1	160	3	15	6.8	11.64	92.98	3.18	10.82
185	0.31	1143	240	0.5	15	16	15.32	70.98	9.87	10.87
186	0.376	1185	320	1	15	6.8	13.06	83.42	3.14	10.89
187	0.121	1169.9	80	1	15	4	14.17	77.79	8.56	11.02
188	0.258	1175	120	2	15	6.8	12.53	88.25	4.73	11.06
189	0.088	1140	80	1	15	6.8	16.04	69.22	12.27	11.1
190	0.121	1145	80	1	30	4	14.9	74.68	9.49	11.12

Table S1. (Continued)

Index	Pressure (barg)	Temperature (°C)	Flow rate (sccm)	H_2 content	Reactor length (cm)	Reactor diameter (mm)	CH_4 conversion (%)	C_2 selectivity (%)	Coke selectivity (%)	C_2 yield (%)
191	0.156	1165	80	3	15	6.8	13.77	80.9	8.86	11.14
192	0.193	1197.5	240	5	15	16	12.56	89.23	5.9	11.21
193	0.089	1140	80	1	15	6.8	16.83	66.91	13.83	11.26
194	0.119	1140	80	1	15	6.8	15.77	71.45	8.82	11.27
195	0.112	1145.1	80	2	45	4	13.41	84.04	8.49	11.27
196	0.089	1140	80	1	15	6.8	17.3	65.79	14.63	11.38
197	0.089	1140	80	1	15	6.8	17.44	65.59	14.34	11.44
198	0.091	1140	80	1	15	6.8	17.93	63.89	9.96	11.46
199	0.091	1140	80	1	15	6.8	18.29	62.83	13.3	11.49
200	0.092	1140	80	1	15	6.8	17.92	64.54	14.4	11.57
201	0.095	1140	80	1	15	6.8	18.39	62.93	14.72	11.57
202	0.092	1140	80	1	15	6.8	17.47	66.26	12.65	11.58
203	0.097	1140	80	1	15	6.8	18.13	63.87	12.98	11.58
204	0.093	1140	80	1	15	6.8	18.14	63.93	14.93	11.6
205	0.095	1140	80	1	15	6.8	17.89	64.97	12.92	11.62
206	0.392	1175	240	1	15	6.8	15.12	77.05	6.88	11.65
207	0.095	1140	80	1	15	6.8	17.9	65.13	13.1	11.66
208	0.476	1160	160	1	15	6.8	16.51	70.69	10.19	11.67
209	0.288	1190.1	160	3	15	6.8	13.53	86.62	9.01	11.72
210	0.193	1200	240	5	15	16	13.98	85.09	9.34	11.9
211	0.259	1180	120	2	15	6.8	14.17	84.23	8.29	11.94
212	0.377	1190	320	1	15	6.8	14.81	80.68	4.14	11.95
213	0.157	1170	80	3	15	6.8	14.73	81.82	6.4	12.06
214	0.12	1145	80	1	15	6.8	18.18	66.32	12.18	12.06
215	0.122	1174.9	80	1	15	4	16.17	74.74	9.72	12.08
216	0.121	1149.9	80	1	30	4	17.1	70.82	11.76	12.11
217	0.112	1149.9	80	2	45	4	15.15	81.38	9.79	12.33
218	0.498	1165	160	1	15	6.8	19.26	64.27	15.28	12.38
219	0.398	1180	240	1	15	6.8	17.48	71.83	10.34	12.56
220	0.641	1170	160	1	15	6.8	24.03	52.64	24.61	12.65
221	0.288	1195.1	160	3	15	6.8	13.93	92.13	2.17	12.84
222	0.119	1149.9	80	1	15	6.8	20.46	63.09	14.38	12.91
223	0.379	1195	320	1	15	6.8	17.47	73.92	9.82	12.91
224	0.26	1185	120	2	15	6.8	15.3	84.6	6.78	12.95
225	0.158	1175	80	3	15	6.8	16.4	79.16	11.74	12.98
226	0.121	1155.1	80	1	30	4	19.55	66.8	14.06	13.06
227	0.122	1180	80	1	15	4	18.55	70.95	11.85	13.16
228	0.119	1155	80	1	15	6.8	23.19	58.15	18.13	13.49
229	0.112	1155	80	2	45	4	17.01	79.34	10.69	13.5
230	0.423	1185	240	1	15	6.8	19.93	67.78	12.39	13.51
231	0.836	1195	240	1	15	6.8	29.98	45.81	31.73	13.74
232	0.26	1190	120	2	15	6.8	16.66	83.27	9.31	13.88
233	0.119	1159.9	80	1	15	6.8	25.98	53.42	22.67	13.88
234	0.122	1159.9	80	1	30	4	22.04	62.99	16.88	13.88
235	0.158	1180	80	3	15	6.8	17.8	78.35	8.4	13.95
236	0.385	1200	320	1	15	6.8	18.94	74.28	6.8	14.06
237	0.12	1165	80	1	15	6.8	28.95	49	27.41	14.18
238	0.525	1190	240	1	15	6.8	23.52	60.83	17.32	14.31
239	0.121	1169.9	80	1	15	6.8	31.87	44.93	32.15	14.32
240	0.122	1174.9	80	1	15	6.8	34.89	41.14	37.06	14.35
241	0.112	1160	80	2	45	4	19.12	76.77	11.89	14.68
242	0.121	1165	80	1	30	4	24.63	59.83	18.86	14.73
243	0.158	1185	80	3	15	6.8	19.48	75.84	9.77	14.77
244	0.261	1195	120	2	15	6.8	18.33	80.67	8.89	14.79
245	0.112	1175	80	2	45	4	18.67	80.39	9.54	15.01
246	0.121	1169.9	80	1	30	4	27.54	55.44	22.83	15.27
247	0.112	1165	80	2	45	4	20.77	75.24	12.09	15.63
248	0.261	1200	120	2	15	6.8	20.63	76.06	14.38	15.69
249	0.159	1190	80	3	15	6.8	20.85	75.31	10.36	15.7
250	0.121	1174.9	80	1	30	4	30.9	50.95	27.69	15.74

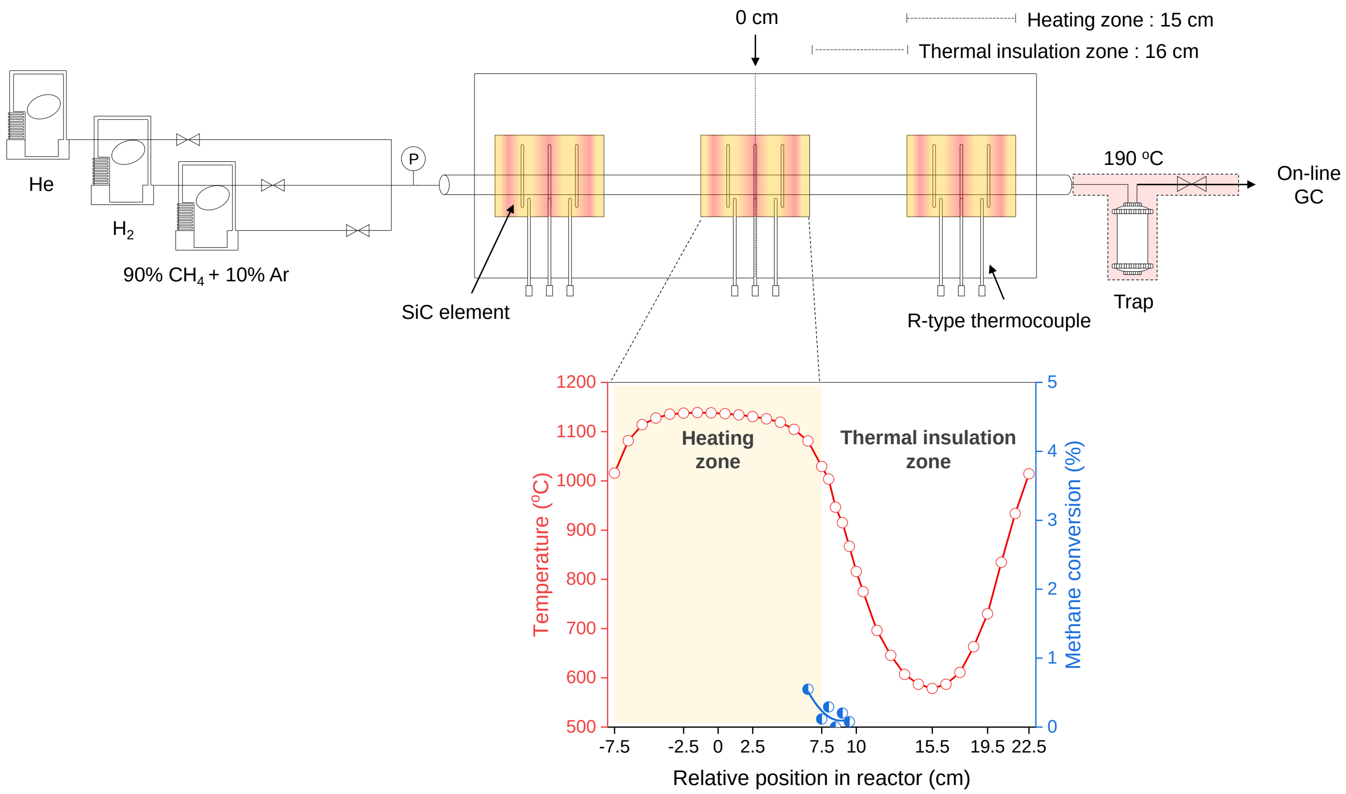

Fig. S1. Schematic diagram of the reactor system. The temperature profile was obtained by setting the heating zone to 1140 °C at a flow rate of 50 mL min^{-1} using He. The reactivity in the thermal insulation zone was measured assuming isothermal using a feed with a ratio of $H_2/(CH_4 + Ar) = 1$ at 2546 h^{-1} of space velocity.

Table S2. Tested hyperparameters for constructing FNN and XGB models. Considering the efficiency and accuracy, we selected these range of parameters. We checked all possible combinations of listed parameters.

Method	Hyperparameters
FNN	Number of nodes: 10, 20, 30, 40, 50, 60, 70 Number of hidden layers: 1, 2, 3, 4, 5 Optimizer: Adam, Adadelta, RMSProp Activation function: ReLU, Sigmoid, Tanh
XGB	Depth: 3, 4, 5, 6, 7, 8, 9 Number of estimators: 100, 200, 300, 400, 500, 600, 700

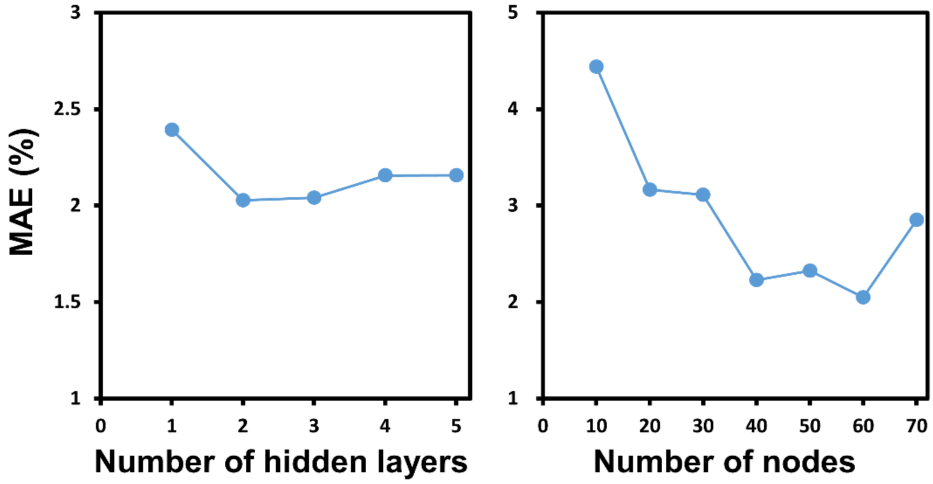

Figure S2. Hyperparameter optimization for FNN. MAE values are exhibited according to the hidden layer (left) and the nodes on this layer (right). We used the Adadelta optimizer and Sigmoid activation function to see the convergence behavior due to the increased number of nodes.

Table S3. Indices for test sets, which are used for constructing FNN and XGB models to optimize hyperparameters and C_2 yield. The numbering index is the same as in the Table S1.

Index	6, 9, 16, 23, 24, 27, 30, 37, 38, 39, 46, 49, 54, 55, 58, 65, 71, 85, 98, 103, 106, 109, 112, 117, 118, 119, 127, 128, 141, 150, 154, 155, 157, 158, 164, 168, 170, 184, 185, 188, 200, 202, 213, 214, 218, 221, 224, 231, 249, 250

Mathematical details of artificial bee colony (ABC) algorithm. The ABC algorithm is a population-based metaheuristic optimization. In our case, the population means a group of possible reaction conditions for non-oxidative conversion of methane. The ABC algorithm tries to find the best solution that minimizes or maximizes a given objective function $L: \mathcal{X} \to \mathbb{R}$ from a randomly distributed populations, $X^{(0)} = \left\{\mathbf{x}_1^{(0)}, \mathbf{x}_2^{(0)}, \dots, \mathbf{x}_N^{(0)}\right\}$, where $\mathcal{X}$ is domain of feasible solutions, and $\mathbf{x}_i^{(k)} \in \mathbb{R}^d$ is a solution vector that contains all condition variables. In our case, we need to find the best solution vector with reaction conditions such as temperature and pressure that will give the best reaction result. In an iterative search of the ABC algorithm, a new candidate solution $\mathbf{v}_i^{(k)} \in \mathbb{R}^d$ is generated by:

$$\mathbf{v}_{ij}^{(k)} = \mathbf{x}_{ij}^{(k-1)} + \alpha_{ij}\left(\mathbf{x}_{ij}^{(k-1)} - \mathbf{x}_{pj}^{(k-1)}\right),$$

where $\mathbf{x}_p^{(k-1)}$ is a randomly selected candidate solution from the population $X^{(k-1)}$, $\alpha_{ij} \in \mathbb{R}$ is a uniform or Gaussian random number within [-1,1], and j is a random dimension index selected from the set of integers {1,2, … , d}. Here, $\mathbf{v}_{ij}^{(k)}$ and $\mathbf{x}_{ij}^{(k)}$ indicate j^{th} elements of $\mathbf{v}_i^{(k)}$ and $\mathbf{x}_i^{(k)}$, respectively. Once the candidate solution $\mathbf{v}_i^{(k)}$ is generated, a greedy selection is used to replace the previous solution, $\mathbf{x}_i^{(k-1)}$. For a maximization problem, if $L(\mathbf{v}_i^{(k)})$ is greater than $L(\mathbf{x}_i^{(k-1)})$, then $\mathbf{v}_i^{(k)}$ becomes a temporary solution $\mathbf{s}_i^{(k)}$; otherwise $\mathbf{s}_i^{(k)}$ is unchanged as $\mathbf{x}_i^{(k-1)}$. After generating a set of temporary solutions, $S^{(k)} = \left\{\mathbf{s}_1^{(k)}, \mathbf{s}_2^{(k)}, \dots, \mathbf{s}_N^{(k)}\right\}$, the current solutions are probabilistically selected based on a selection mechanism according to a selection probability $P_i^{(k)}$ for each temporary solution. For a maximization problem, this selection probability $P_i^{(k)}$ can be defined as

$$P_i^{(k)} = \frac{L(\mathbf{s}_i^{(k)})}{\sum_{n=1}^{N} L(\mathbf{s}_n^{(k)})} \ .$$

As shown in the definition, the larger $L(\mathbf{s}_i^{(k)})$, the higher the selection probability $P_i^{(k)}$ of $\mathbf{s}_i^{(k)}$. The probabilistic selection based on the selection probability $P_i^{(k)}$ is repeated N times for each $i = \{1, 2, \dots, N\}$ to generate a current population of solutions $X^{(k)} = \left\{\mathbf{x}_1^{(k)}, \mathbf{x}_2^{(k)}, \dots, \mathbf{x}_N^{(k)}\right\}$, where $\mathbf{x}_i^{(k)}$ is a probabilistically selected candidate solution from $S^{(k)} = \left\{\mathbf{s}_1^{(k)}, \mathbf{s}_2^{(k)}, \dots, \mathbf{s}_N^{(k)}\right\}$. If the i^{th} solultion in the current population $X^{(k)}$ cannot be improved over a predefined number, the i^{th} solution is replaced by a new randomly generated solution as:

$$\mathbf{x}_{ij}^{(k)} = lb_j + \alpha_{ij}\left(ub_j - lb_j\right),$$

where lb_j and ub_j are lower and upper bounds of the j^{th} condition variable, respectively. In the ABC algorithm, searching candidate solutions with a probabilistic selection is repeated over a predefined number of iterations to find an optimal solution.

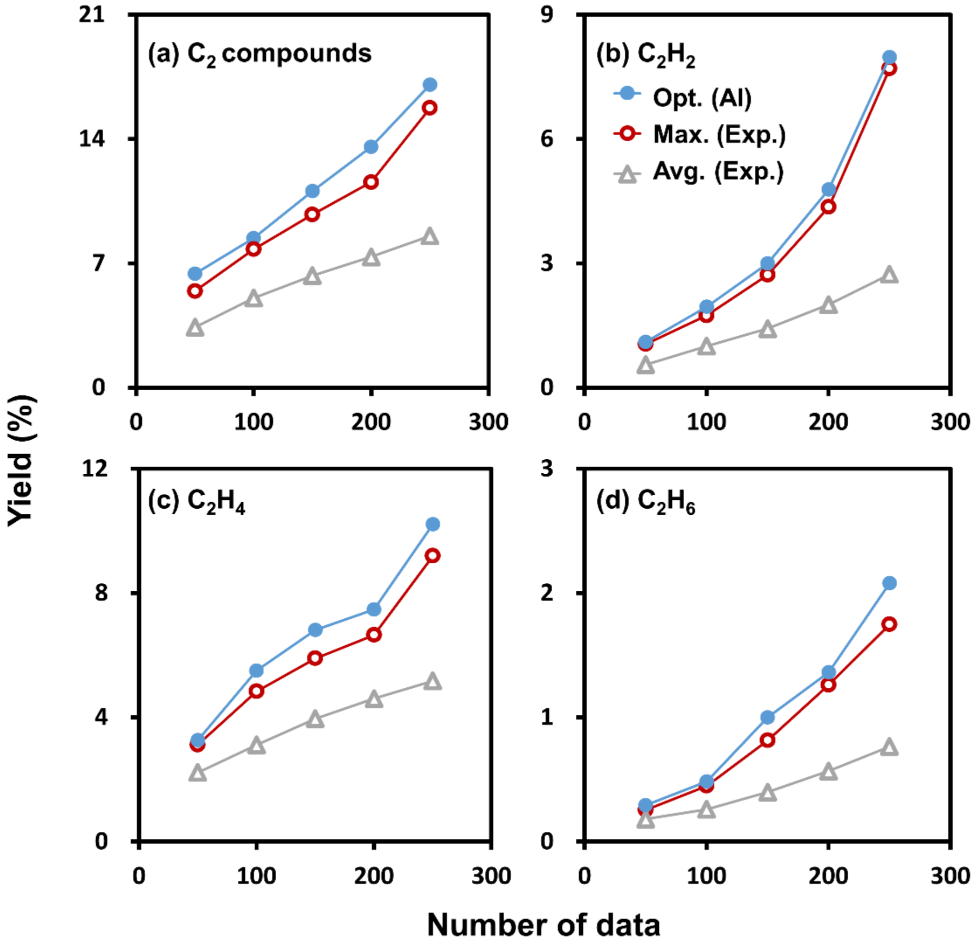

Figure S3. Results of the AI-based optimization for the yield of (a) all C_2 compound, (b) C_2H_2, (c) C_2H_4, and (d) C_2H_6. The AI optimization results (filled blue circles) are compared to the maximum value of the experimental results (open red circles) and their average (open gray triangles).

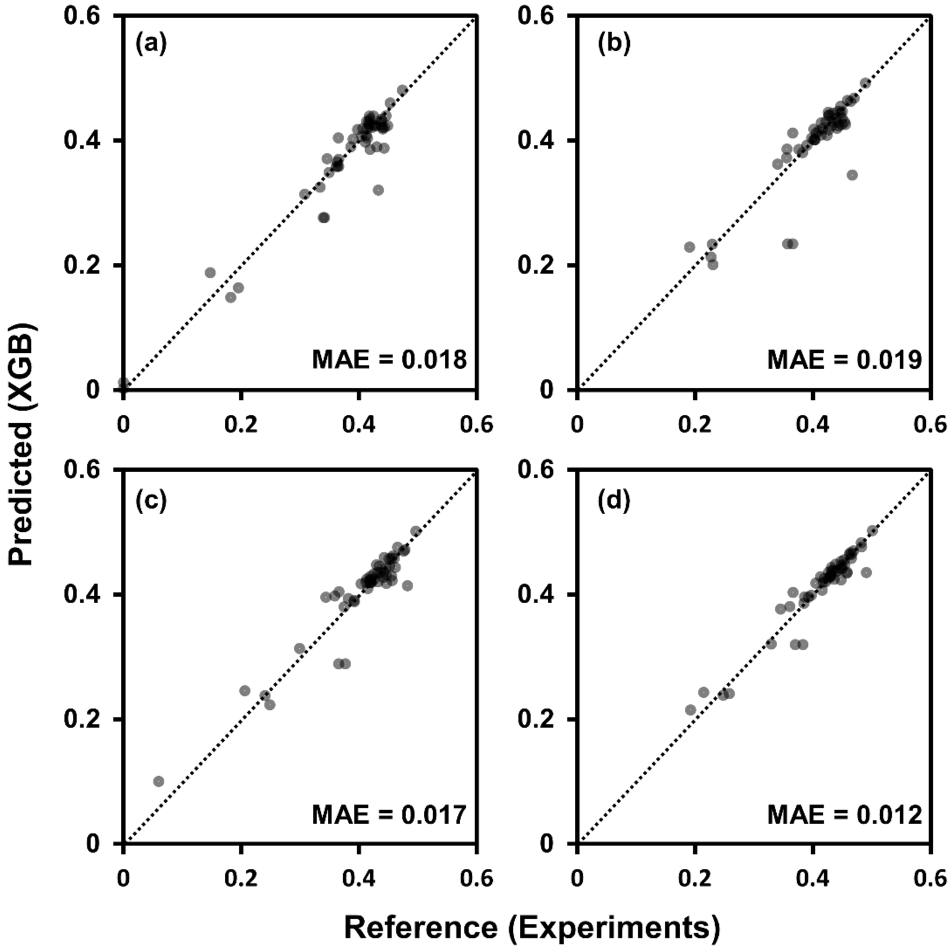

Figure S4. XGB predictions of scoring functions compared to experimental references with cutoff values for the coke: (a) 40%, (b) 60%, (c) 80%, and 100%.

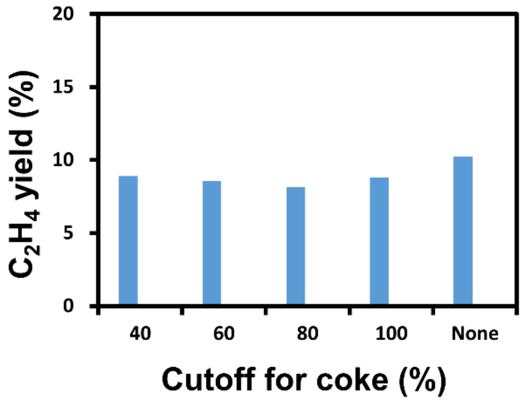

Figure S5. Effects of the cutoff for the C_2H_4 yield.

Table S4. Experimental data, whose C_2 yield and coke selectivity are similar to the experimental validation. We use the same index as Table S1.

Index	Pressure (barg)	Temperature (ºC)	Flow rate (sccm)	H_2 content	Reactor length (cm)	Reactor diameter (mm)	CH_4 conversion (%)	C_2 selectivity (%)	Coke selectivity (%)	C_2 yield (%)
172	0.156	1160	80	3	15	6.8	11.91	86.15	3.6	10.26
175	0.119	1134.9	80	1	15	6.8	13.57	76.33	3.81	10.36
184	0.288	1185.1	160	3	15	6.8	11.64	92.98	3.18	10.82
186	0.376	1185	320	1	15	6.8	13.06	83.42	3.14	10.89
188	0.258	1175	120	2	15	6.8	12.53	88.25	4.73	11.06