

Thermal Activation of Methane by MgO^+ : Temperature Dependent Kinetics, Reactive Molecular Dynamics Simulations and Statistical Modeling

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I. QUALITY OF THE NN-LEARNED FORCE FIELD

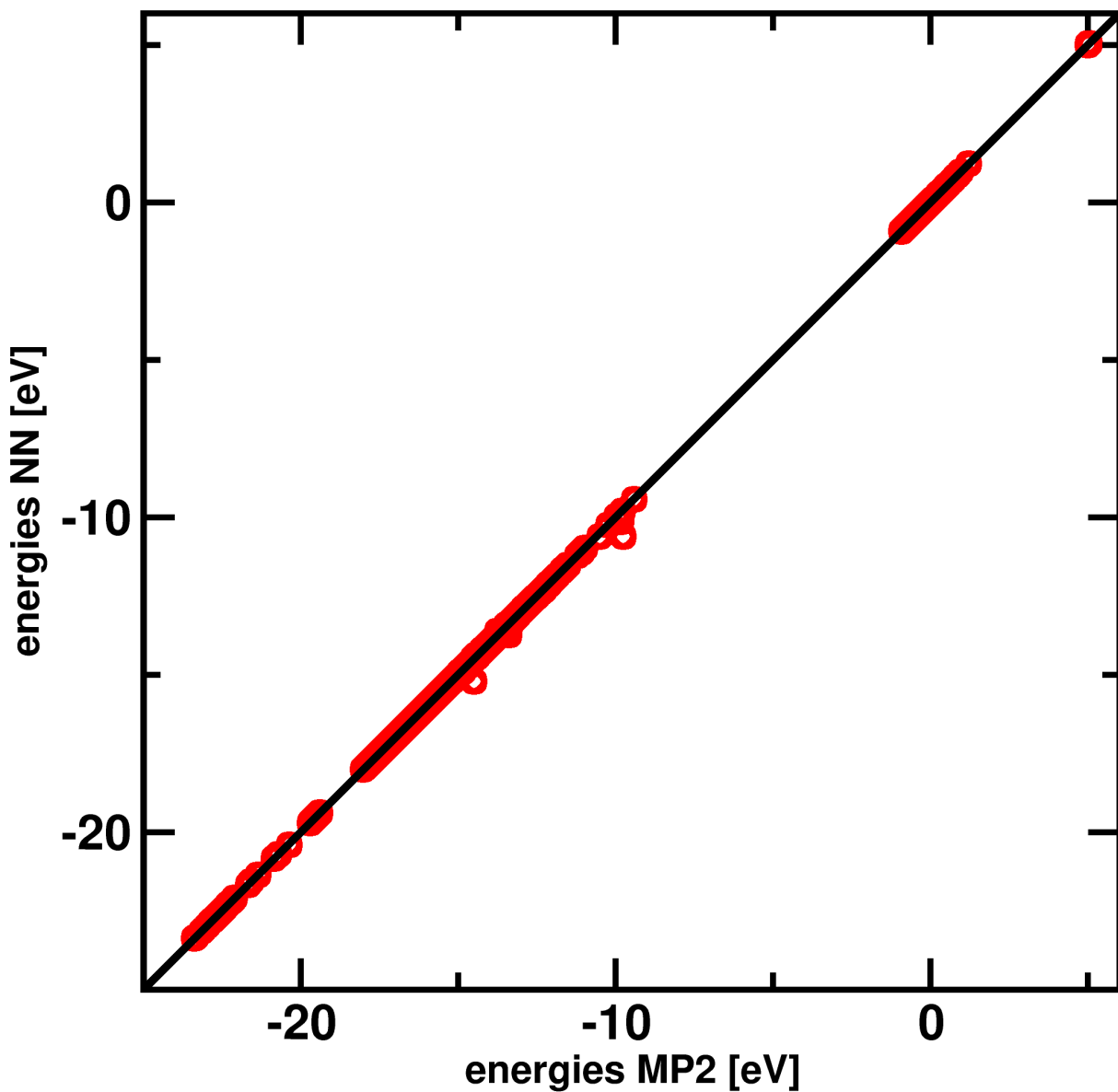


FIG. S1. Correlation between the energies for 4368 *ab initio* reference structures from the test set and the trained neural network. Including the separated molecules (MgO^+ , CH_4 , MgOH^+ , CH_3 , CH_3OH , Mg^+), transition states and complexes with $R^2 = 1 - 4.1 \times 10^{-6}$.

TABLE S1. Summary for all trajectories run on the MS-ARMD PES.

	T [K]	b [Å]	Reactive	Complex	Nonreactive	Total
1	300	0.0–0.5	70	15	688	773
2		0.5–1.0	42	6	448	496
3		1.0–1.5	73	9	729	811
4		1.5–2.0	95	14	903	1012
5		2.0–2.5	22	5	242	269
6		2.5–3.0	7	1	98	106
7		3.0–3.5	6	1	26	33
8	400	0.0–0.5	74	14	799	897
9		0.5–1.0	83	24	1063	1170
10		1.0–1.5	50	9	1010	1069
11		1.5–2.0	9	1	110	120
12		2.0–2.5	12	1	105	118
13		2.5–3.0	7	4	97	108
14		3.0–3.5	0	1	17	18
15	500	0.0–0.5	117	40	964	1121
16		0.5–1.0	93	24	844	961
17		1.0–1.5	43	14	533	590
18		1.5–2.0	29	11	409	449
19		2.0–2.5	14	5	208	227
20		2.5–3.0	2	2	103	107
21		3.0–3.5	2	2	41	45
22	600	0.0–0.5	154	30	1230	1414
23		0.5–1.0	114	23	552	689
24		1.0–1.5	52	8	485	545
25		1.5–2.0	38	10	468	516
26		2.0–2.5	23	7	184	214
27		2.5–3.0	2	2	86	90
28		3.0–3.5	1	0	31	32

TABLE S2. Summary for all trajectories, 500 ps in length, from uniform sampling of the NN PES.

	$T[\text{K}]$	$b \text{ [\AA]}$	Reactive	Complex	Nonreactive	Total
1	300	0.0–0.5	82	1140	73	1295
2		0.5–1.0	82	680	24	786
3		1.0–1.5	9	474	55	538
4		1.5–2.0	6	342	38	386
5		2.0–2.5	0	247	17	264
6		2.5–3.0	0	137	29	166
7		3.0–3.5	0	31	34	65
8	400	0.0–0.5	45	910	342	1297
9		0.5–1.0	31	583	166	780
10		1.0–1.5	17	340	140	497
11		1.5–2.0	10	281	134	425
12		2.0–2.5	13	162	68	243
13		2.5–3.0	5	96	71	172
14		3.0–3.5	0	38	48	86
15	500	0.0–0.5	210	643	425	1278
16		0.5–1.0	61	434	319	814
17		1.0–1.5	42	293	219	554
18		1.5–2.0	22	186	172	380
19		2.0–2.5	12	99	136	247
20		2.5–3.0	7	53	95	155
21		3.0–3.5	18	20	34	72
22	600	0.0–0.5	176	434	712	1322
23		0.5–1.0	101	283	399	783
24		1.0–1.5	71	150	327	548
25		1.5–2.0	23	88	257	368
26		2.0–2.5	15	61	172	248
27		2.5–3.0	7	39	125	171
28		3.0–3.5	3	10	47	60

TABLE S3. Summary for all trajectories, 50 ps in length, from stratified sampling of the NN PES for $T = 300$ K to $T = 500$ K.

	T [K]	b [Å]	Reactive	Complex	Nonreactive	Total
1	300	0.0–0.5	3	1394	180	1577
2		0.5–1.0	1	1492	188	1681
3		1.0–1.5	1	1435	164	1600
4		1.5–2.0	0	1297	127	1424
5		2.0–2.5	0	1122	143	1265
6		2.5–3.0	0	888	206	1094
7		3.0–3.5	0	521	420	941
8		3.5–4.0	0	172	623	795
9		4.0–4.5	0	17	631	648
10		4.5–5.0	0	1	476	477
11		5.0–5.5	0	0	294	294
12		5.5–6.0	0	0	167	167
13	400	0.0–0.5	11	1291	195	1497
14		0.5–1.0	9	1326	268	1603
15		1.0–1.5	8	1152	242	1402
16		1.5–2.0	2	982	225	1209
17		2.0–2.5	1	812	182	995
18		2.5–3.0	0	560	261	821
19		3.0–3.5	0	261	379	640
20		3.5–4.0	0	48	387	435
21		4.0–4.5	0	2	191	193
22	500	0.0–0.5	25	1054	430	1509
23		0.5–1.0	19	1121	469	1609
24		1.0–1.5	13	993	359	1365
25		1.5–2.0	5	792	311	1108
26		2.0–2.5	3	582	322	907
27		2.5–3.0	0	406	294	700
28		3.0–3.5	1	145	259	405
29		3.5–4.0	0	66	257	323

TABLE S4. Summary for all trajectories, 50 ps in length, from stratified sampling of the NN PES for $T = 600$ K and $T = 700$ K.

	T [K]	b [Å]	Reactive	Complex	Nonreactive	Total
30	600	0.0–0.5	53	891	551	1495
31		0.5–1.0	38	991	528	1557
32		1.0–1.5	16	811	542	1369
33		1.5–2.0	8	622	525	1155
34		2.0–2.5	6	515	405	926
35		2.5–3.0	1	306	407	714
36		3.0–3.5	1	92	355	448
37		3.5–4.0	0	16	238	254
38	700	0.0–0.5	64	697	782	1543
39		0.5–1.0	58	685	823	1566
40		1.0–1.5	27	621	733	1381
41		1.5–2.0	16	484	639	1139
42		2.0–2.5	7	354	552	913
43		2.5–3.0	4	209	509	722
44		3.0–3.5	1	71	430	502
45		3.5–4.0	0	7	195	202