

# Atomistic Molecular Dynamics Simulations of H<sub>2</sub>O Diffusivity in Liquid and Supercritical CO<sub>2</sub>

## Supplemental Information

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# 1. Self-diffusion coefficient of pure CO<sub>2</sub> and diffusion coefficient of H<sub>2</sub>O in CO<sub>2</sub> at infinite dilution

Table S1. TIP4P/2005 – EPM2 model

Temperature (K)	Pressure (MPa)	Density (kg m <sup>-3</sup> )	Self-Diffusion coefficient of CO <sub>2</sub> for a system with 1000 molecules (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )	Self-Diffusion coefficient of CO <sub>2</sub> extrapolated to infinite system size (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )	Diffusion coefficient of H <sub>2</sub> O in CO <sub>2</sub> (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )
283.15	5	840 ± 5	16.2 ± 0.3	17.8 ± 0.3	19 ± 2
	10	898 ± 4	14.0 ± 0.2	15.4 ± 0.2	15 ± 2
	20	962 ± 4	11.9 ± 0.2	13.1 ± 0.2	14 ± 2
	30	1002 ± 3	10.7 ± 0.3	11.8 ± 0.2	13 ± 2
	50	1060 ± 3	9.0 ± 0.1	9.9 ± 0.1	12 ± 2
298.15	7	229.3 ± 0.5	90.3 ± 0.9	122 ± 3	104 ± 10
	10	780 ± 5	19.6 ± 0.4	21.4 ± 0.4	22 ± 3
	13	819 ± 4	17.8 ± 0.4	19.5 ± 0.5	20 ± 4
	16	853 ± 4	16.4 ± 0.3	18.0 ± 0.4	19 ± 3
	20	892 ± 4	15.0 ± 0.3	16.5 ± 0.3	17 ± 3
	30	948 ± 3	12.9 ± 0.2	14.2 ± 0.3	15 ± 3
	50	1017 ± 3	10.9 ± 0.2	12.0 ± 0.2	13 ± 2
308.15	7	187.2 ± 0.3	104 ± 2	134 ± 3	132 ± 13
	10	667 ± 8	25.5 ± 0.5	31 ± 1	32 ± 5
	13	742 ± 5	22.0 ± 0.3	24.1 ± 0.6	28 ± 5
	16	803 ± 4	19.1 ± 0.4	21.0 ± 0.3	22 ± 3
	20	840 ± 4	17.7 ± 0.3	19.4 ± 0.4	21 ± 3
	30	909 ± 3	15.0 ± 0.3	16.5 ± 0.3	19 ± 3
	50	986 ± 3	12.3 ± 0.3	13.5 ± 0.3	16 ± 2
323.15	10	300 ± 2	68.5 ± 0.7	83 ± 3	77 ± 6
	20	744 ± 4	22.7 ± 0.4	24.8 ± 0.5	26 ± 3
	30	850 ± 3	17.8 ± 0.4	19.6 ± 0.4	23 ± 3
	50	949 ± 3	14.0 ± 0.2	15.5 ± 0.3	17 ± 3
	100	1066 ± 2	10.4 ± 0.2	11.5 ± 0.2	13 ± 2
423.15	10	141.3 ± 0.2	170 ± 3	262 ± 8	202 ± 13
	20	311.8 ± 0.6	81 ± 1	104 ± 3	92 ± 10
	30	463 ± 2	52 ± 1	65 ± 2	68 ± 8
	50	663 ± 2	33.6 ± 0.7	45 ± 3	45 ± 6
	100	873 ± 2	21.2 ± 0.4	23.4 ± 0.4	26 ± 4
523.15	10	101.3 ± 0.2	240 ± 5	457 ± 12	295 ± 18
	20	208.9 ± 0.4	126 ± 3	207 ± 5	143 ± 12
	30	311.9 ± 0.7	88 ± 2	135 ± 4	108 ± 8
	50	479 ± 1	57 ± 1	73 ± 2	69 ± 9
	100	722 ± 2	34.6 ± 0.7	43 ± 2	39 ± 4

Table S2. TIP4P/2005 – TraPPE model

Temperature (K)	Pressure (MPa)	Density (kg m <sup>-3</sup> )	Self-Diffusion coefficient of CO <sub>2</sub> for a system with 1000 molecules (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )	Self-Diffusion coefficient of CO <sub>2</sub> extrapolated to infinite system size (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )	Diffusion coefficient of H <sub>2</sub> O in CO <sub>2</sub> (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )
283.15	5	882 ± 5	13.5 ± 0.3	15.5 ± 0.3	15 ± 2
	10	924 ± 4	12.2 ± 0.2	13.6 ± 0.4	14 ± 2
	20	975 ± 3	10.7 ± 0.2	11.9 ± 0.6	12 ± 2
	30	1011 ± 3	9.6 ± 0.2	10.7 ± 0.3	11 ± 2
	50	1063 ± 3	8.2 ± 0.1	9.1 ± 0.1	10 ± 2
298.15	7	757 ± 6	18.7 ± 0.3	21.7 ± 0.8	21 ± 3
	10	822 ± 5	16.6 ± 0.4	18.5 ± 0.5	19 ± 3
	13	860 ± 4	15.2 ± 0.3	16.9 ± 0.4	18 ± 3
	16	884 ± 4	14.3 ± 0.3	15.9 ± 0.4	17 ± 3
	20	910 ± 4	13.4 ± 0.2	14.9 ± 0.3	16 ± 3
	30	958 ± 3	11.7 ± 0.2	13.0 ± 0.3	14 ± 2
	50	1021 ± 3	9.9 ± 0.2	11.0 ± 0.2	13 ± 3
308.15	7	169.7 ± 0.5	110 ± 2	178 ± 3	110 ± 13
	10	727 ± 5	21.6 ± 0.3	23.5 ± 0.9	23 ± 4
	13	789 ± 4	18.7 ± 0.3	20.8 ± 0.4	20 ± 3
	16	827 ± 4	17.2 ± 0.3	19.1 ± 0.4	19 ± 4
	20	862 ± 4	15.7 ± 0.3	17.4 ± 0.5	18 ± 3
	30	920 ± 3	13.5 ± 0.2	15.0 ± 0.3	17 ± 3
	50	992 ± 3	11.1 ± 0.2	12.6 ± 0.4	13 ± 2
323.15	10	281 ± 1	70 ± 1	117 ± 2	74 ± 8
	20	770 ± 4	20.4 ± 0.4	23.0 ± 0.8	23 ± 3
	30	856 ± 3	16.6 ± 0.3	18.4 ± 0.4	19 ± 2
	50	945 ± 3	13.4 ± 0.3	14.9 ± 0.3	16 ± 2
	100	1061 ± 2	9.6 ± 0.2	10.7 ± 0.3	13 ± 2
423.15	10	135.8 ± 0.3	167 ± 3	271 ± 2	176 ± 19
	20	305.2 ± 0.8	79 ± 2	108 ± 2	101 ± 16
	30	467 ± 2	50.6 ± 0.8	64 ± 1	65 ± 8
	50	664 ± 2	32.4 ± 0.7	40.3 ± 0.6	39 ± 6
	100	869 ± 2	20.6 ± 0.5	23.1 ± 0.5	26 ± 3
473.15	10	116.2 ± 0.2	200 ± 4	358 ± 3	229 ± 20
	20	245.6 ± 0.6	103 ± 2	149 ± 3	122 ± 16
	100	786 ± 0.2	26.9 ± 0.6	29.7 ± 0.5	39 ± 4
523.15	10	101.9 ± 0.2	232 ± 4	423 ± 6	269 ± 23
	20	209.5 ± 0.4	123 ± 2	201 ± 6	144 ± 22
	30	311.7 ± 0.7	86 ± 2	134 ± 2	103 ± 14
	50	482 ± 1	55.2 ± 0.9	80 ± 1	69 ± 8
	100	717 ± 2	33.2 ± 0.5	43 ± 1	41 ± 5

Table S3. TIP4P/2005 – ZD model

Temperature (K)	Pressure (MPa)	Density (kg m <sup>-3</sup> )	Self-Diffusion coefficient of CO <sub>2</sub> for a system with 1000 molecules (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )	Self-Diffusion coefficient of CO <sub>2</sub> extrapolated to infinite system size (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )	Diffusion coefficient of H <sub>2</sub> O in CO <sub>2</sub> (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )
283.15	5	816 ± 6	17.8 ± 0.4	19.4 ± 0.4	19 ± 2
	10	887 ± 4	15.1 ± 0.3	16.5 ± 0.4	16 ± 2
	20	956 ± 4	12.7 ± 0.3	13.9 ± 0.3	15 ± 2
	30	1000 ± 3	11.4 ± 0.3	12.5 ± 0.3	14 ± 2
	50	1060 ± 3	9.8 ± 0.2	10.7 ± 0.2	12 ± 2
298.15	7	189.4 ± 0.7	101 ± 2	144 ± 5	113 ± 11
	10	773 ± 5	20.7 ± 0.3	22.5 ± 0.5	26 ± 4
	13	824 ± 4	18.4 ± 0.4	20.1 ± 0.4	23 ± 4
	16	856 ± 4	17.1 ± 0.3	18.7 ± 0.4	20 ± 3
	20	890 ± 4	15.8 ± 0.3	17.3 ± 0.3	19 ± 2
	30	947 ± 3	13.8 ± 0.3	15.1 ± 0.3	17 ± 2
	50	1019 ± 3	11.4 ± 0.2	12.5 ± 0.3	15 ± 3
308.15	7	171.0 ± 0.6	114 ± 2	180 ± 4	131 ± 17
	10	570 ± 8	33.1 ± 0.6	42 ± 2	35 ± 5
	13	739 ± 5	22.9 ± 0.6	25.0 ± 0.8	25 ± 4
	16	791 ± 4	20.5 ± 0.4	22.4 ± 0.5	24 ± 3
	20	837 ± 4	18.2 ± 0.3	19.9 ± 0.4	19 ± 3
	30	908 ± 3	15.5 ± 0.3	17.0 ± 0.3	18 ± 3
	50	990 ± 3	12.9 ± 0.5	14.1 ± 0.2	15 ± 3

**Table S4. TIP4P/2005 – Merker et al. model**

Temperature (K)	Pressure (MPa)	Density (kg m <sup>-3</sup> )	Self-Diffusion coefficient of CO <sub>2</sub> for a system with 1000 molecules (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )	Self-Diffusion coefficient of CO <sub>2</sub> extrapolated to infinite system size (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )	Diffusion coefficient of H <sub>2</sub> O in CO <sub>2</sub> (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )
283.15	5	857 ± 5	13.8 ± 0.3	15.4 ± 0.3	16 ± 2
	10	903 ± 4	12.1 ± 0.3	13.5 ± 0.3	15 ± 3
	20	959 ± 4	10.6 ± 0.3	11.8 ± 0.3	13 ± 3
	30	997 ± 3	9.4 ± 0.2	10.5 ± 0.3	11 ± 2
	50	1051 ± 3	8.1 ± 0.2	9.0 ± 0.2	10 ± 3
298.15	7	743 ± 6	19.0 ± 0.2	21.2 ± 0.7	23 ± 4
	10	804 ± 4	16.6 ± 0.5	18.5 ± 0.4	19 ± 3
	13	839 ± 5	15.2 ± 0.3	16.9 ± 0.5	18 ± 3
	16	865 ± 4	14.3 ± 0.3	15.9 ± 0.4	17 ± 4
	20	893 ± 3	13.2 ± 0.2	14.7 ± 0.3	16 ± 4
	30	943 ± 4	11.6 ± 0.3	12.9 ± 0.3	14 ± 2
	50	1008 ± 3	9.7 ± 0.2	10.8 ± 0.3	13 ± 3
308.15	7	327 ± 9	49 ± 1	61 ± 3	40 ± 4
	10	700 ± 6	21.8 ± 0.5	24.0 ± 0.6	27 ± 5
	13	768 ± 4	18.6 ± 0.4	20.6 ± 0.5	23 ± 4
	16	807 ± 4	17.0 ± 0.2	18.9 ± 0.4	20 ± 3
	20	844 ± 4	15.4 ± 0.3	17.1 ± 0.2	18 ± 3
	30	905 ± 3	13.2 ± 0.2	14.7 ± 0.3	16 ± 2
	50	979 ± 3	10.9 ± 0.3	12.1 ± 0.2	15 ± 2

Table S5. Exp-6 – Exp-6 model

Temperature (K)	Pressure (MPa)	Density (kg m <sup>-3</sup> )	Self-Diffusion coefficient of CO <sub>2</sub> for a system with 1000 molecules (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )	Self-Diffusion coefficient of CO <sub>2</sub> extrapolated to infinite system size (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )	Diffusion coefficient of H <sub>2</sub> O in CO <sub>2</sub> (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )
283.15	5	857 ± 5	16.1 ± 0.3	17.7 ± 0.4	17 ± 3
	10	913 ± 4	14.0 ± 0.4	15.4 ± 0.4	15 ± 3
	20	976 ± 4	12.0 ± 0.3	13.2 ± 0.3	11 ± 2
	30	1017 ± 3	10.8 ± 0.3	11.9 ± 0.4	10 ± 2
	50	1075 ± 3	9.3 ± 0.2	10.2 ± 0.3	9 ± 2
298.15	7	695 ± 8	24.5 ± 0.6	29.1 ± 0.8	26 ± 5
	10	800 ± 5	19.2 ± 0.3	21.1 ± 0.4	20 ± 4
	13	845 ± 4	17.6 ± 0.4	19.3 ± 0.3	19 ± 5
	16	876 ± 4	16.3 ± 0.3	17.9 ± 0.4	19 ± 4
	20	907 ± 4	15.1 ± 0.3	16.6 ± 0.3	17 ± 3
	30	962 ± 3	13.2 ± 0.3	14.5 ± 0.4	13 ± 2
	50	1032 ± 3	11.0 ± 0.2	12.1 ± 0.3	11 ± 2
308.15	7	181.5 ± 0.6	108 ± 2	175 ± 0.5	119 ± 15
	10	663 ± 6	27.0 ± 0.4	29.9 ± 0.9	28 ± 7
	13	764 ± 5	21.7 ± 0.4	23.7 ± 0.8	23 ± 5
	16	812 ± 4	19.4 ± 0.5	21.3 ± 0.4	24 ± 5
	20	855 ± 4	17.6 ± 0.3	19.3 ± 0.4	16 ± 2
	30	923 ± 3	15.1 ± 0.4	16.6 ± 0.4	15 ± 3
	50	1002 ± 3	12.2 ± 0.3	13.4 ± 0.3	11 ± 2
323.15	10	294 ± 1	71.4 ± 0.2	119 ± 0.3	68 ± 9
	20	767 ± 4	22.2 ± 0.4	24.3 ± 0.6	25 ± 7
	30	861 ± 4	18.1 ± 0.4	19.9 ± 0.4	21 ± 5
	50	958 ± 3	14.4 ± 0.3	15.9 ± 0.4	15 ± 3
	100	1081 ± 3	10.5 ± 0.2	11.6 ± 0.3	12 ± 2
423.15	10	140.2 ± 0.3	166 ± 3	268 ± 9	158 ± 21
	20	311.3 ± 0.8	81 ± 2	105 ± 4	82 ± 9
	30	473 ± 2	52.9 ± 0.9	66 ± 3	51 ± 7
	50	674 ± 2	34.1 ± 0.7	45 ± 2	33 ± 5
	100	887 ± 3	21.6 ± 0.4	23.8 ± 0.5	24 ± 4
473.15	10	118.9 ± 0.2	203 ± 4	402 ± 11	188 ± 20
	20	250.2 ± 0.6	104 ± 2	133 ± 4	108 ± 12
	100	804 ± 2	28.1 ± 0.7	30.9 ± 0.7	26 ± 5
523.15	10	104.1 ± 0.2	238 ± 4	455 ± 9	236 ± 32
	20	213.3 ± 0.5	127 ± 3	210 ± 6	123 ± 15
	30	317.8 ± 0.8	88 ± 2	112 ± 4	91 ± 10
	50	489 ± 1	57 ± 1	75 ± 3	60 ± 10
	100	733 ± 2	34.7 ± 0.8	41 ± 2	31 ± 6

## 2. Maxwell-Stefan diffusion coefficient of H<sub>2</sub>O in CO<sub>2</sub> at various H<sub>2</sub>O compositions

Table S6. TIP4P/2005 – TraPPE model

Temperature (K)	Pressure (MPa)	Density (kg m <sup>-3</sup> )	H <sub>2</sub> O mole fraction	# of H <sub>2</sub> O molecules	D <sub>MS</sub> (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )
473.15	10	116.2 ± 0.2	0.001	1	229 ± 20
		113.7 ± 0.2	0.0476	50	232 ± 22
		111.9 ± 0.2	0.0909	100	275 ± 40
	20	245.6 ± 0.6	0.001	1	122 ± 16
		243.0 ± 0.6	0.0476	50	132 ± 18
		242.4 ± 0.6	0.0909	100	140 ± 23
	100	786 ± 2	0.001	1	39 ± 4
		786 ± 2	0.0476	50	42 ± 6
		787 ± 2	0.0909	100	51 ± 5
523.15	10	101.3 ± 0.2	0.001	1	295 ± 18
		98.1 ± 0.2	0.0476	50	267 ± 32
		95.8 ± 0.2	0.0909	100	316 ± 25
		94.4 ± 0.2	0.1304	150	329 ± 33
		92 ± 1	0.2	250	328 ± 32
	20	208.9 ± 0.4	0.001	1	143 ± 12
		204.8 ± 0.4	0.0476	50	161 ± 20
		201.8 ± 0.4	0.0909	100	181 ± 24
		200.3 ± 0.5	0.1304	150	180 ± 30
	100	722 ± 2	0.001	1	39 ± 4
		712 ± 2	0.0476	50	54 ± 11
		711 ± 2	0.0909	100	49 ± 9

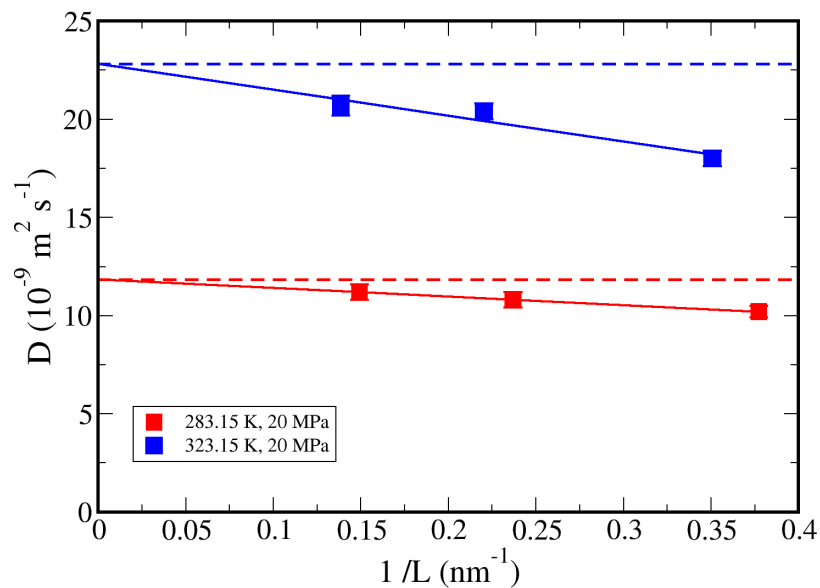
Table S7. Exp-6 – Exp-6 model

Temperature (K)	Pressure (MPa)	Density (kg m <sup>-3</sup> )	H <sub>2</sub> O mole fraction	# of H <sub>2</sub> O molecules	D <sub>MS</sub> (10 <sup>-9</sup> m <sup>2</sup> s <sup>-1</sup> )	
473.15	10	118.9 ± 0.2	0.001	1	188 ± 20	
		116.8 ± 0.2	0.0476	50	280 ± 30	
		115.1 ± 0.2	0.0909	100	326 ± 45	
		113.2 ± 0.2	0.1525	180	353 ± 49	
	20	250.2 ± 0.6	0.001	1	108 ± 12	
		249.9 ± 0.6	0.0476	50	167 ± 20	
		250.9 ± 0.6	0.0909	100	184 ± 24	
		255.5 ± 0.7	0.1525	180	169 ± 21	
	100	804 ± 2	0.001	1	26 ± 5	
		818 ± 2	0.0476	50	46 ± 9	
		833 ± 2	0.0909	100	41 ± 7	
		855 ± 3	0.1525	180	35 ± 6	
	523.15	10	104.1 ± 0.2	0.001	1	236 ± 32
			101.9 ± 0.2	0.0476	50	391 ± 49
			99.9 ± 0.2	0.0909	100	437 ± 56
			98.2 ± 0.2	0.1304	150	438 ± 59
95.4 ± 0.2			0.2	250	398 ± 45	
93.4 ± 0.2			0.2593	350	427 ± 54	
91.7 ± 0.2			0.3103	450	411 ± 60	
90.5 ± 0.2			0.3548	550	378 ± 46	
20		213.3 ± 0.5	0.001	1	123 ± 15	
		210.9 ± 0.5	0.0476	50	225 ± 21	
		209.5 ± 0.4	0.0909	100	228 ± 36	
		208.4 ± 0.4	0.1304	150	201 ± 22	
		208.1 ± 0.5	0.2	250	219 ± 35	
		209.7 ± 0.5	0.2593	350	200 ± 38	
		212.8 ± 0.5	0.3103	450	195 ± 23	
		217.5 ± 0.6	0.3548	550	179 ± 21	
100		733 ± 2	0.001	1	31 ± 6	
		742 ± 2	0.0476	50	59 ± 17	
		752 ± 2	0.0909	100	56 ± 11	
		763 ± 2	0.1304	150	55 ± 10	
		785 ± 2	0.2	250	48 ± 9	
		806 ± 2	0.2593	350	43 ± 7	
		826 ± 3	0.3103	450	41 ± 8	



Figure S1. Self-diffusion coefficient of TraPPE CO<sub>2</sub> at (a) 283.15 K and 323.15 K (20 MPa for both temperatures) and (b) 523.15 K (10 and 50 MPa) for various box sizes. Solid lines are the linear fits to the MD values and dashed line shows the extrapolated value at infinite system size.

(a)



(b)

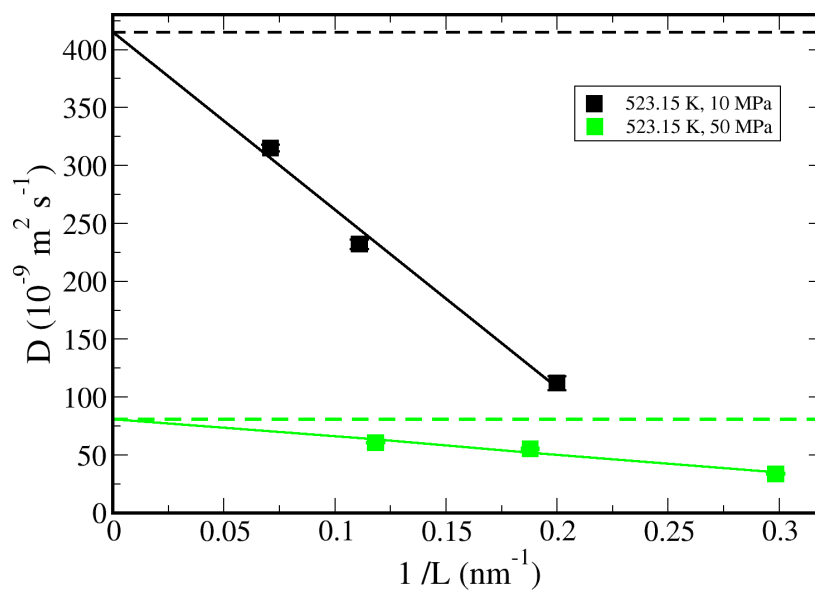


Figure S2. Self-diffusion coefficient of EPM2, TraPPE and ZD CO<sub>2</sub> at 298.15 K as a function of density.

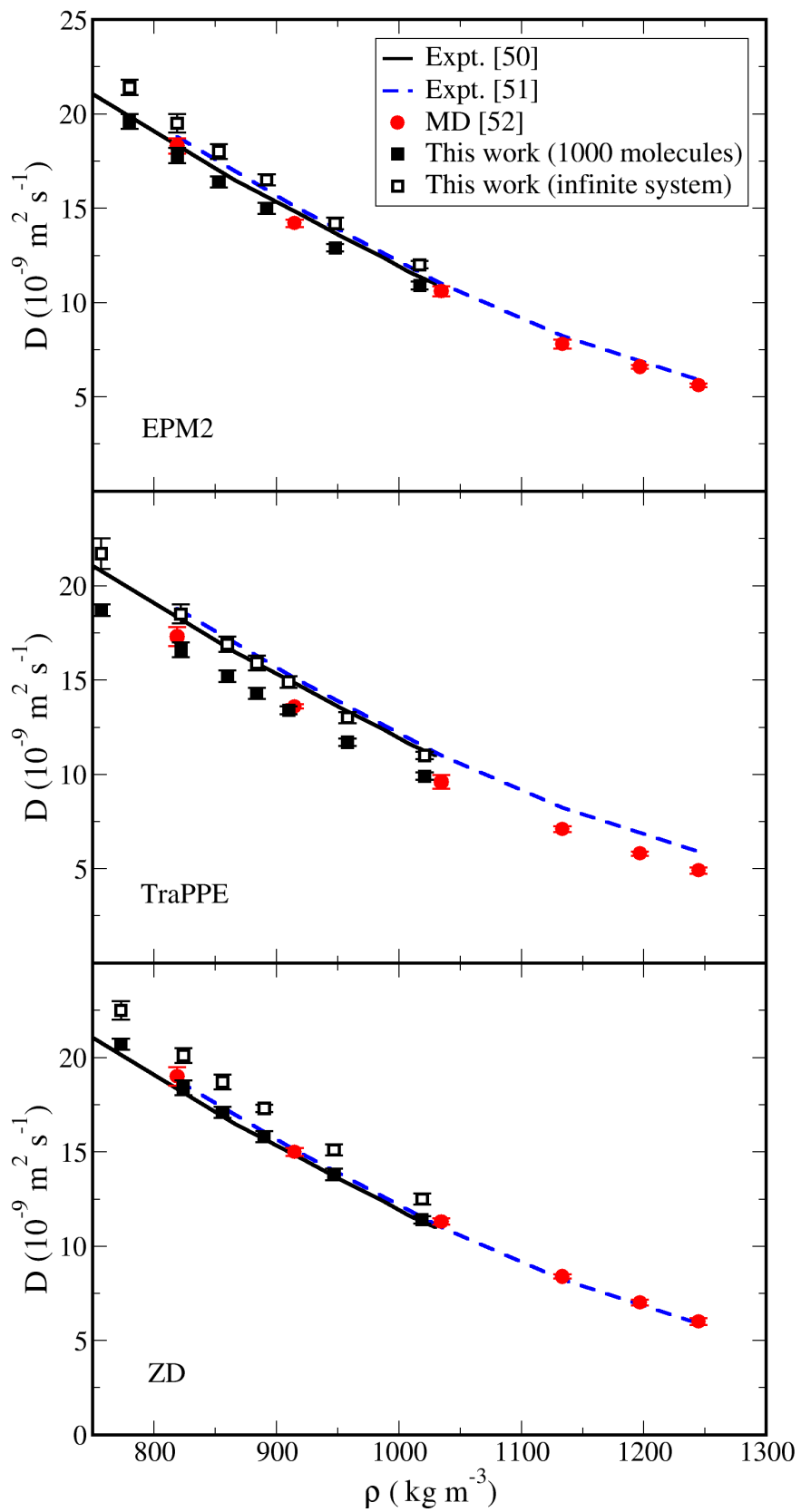
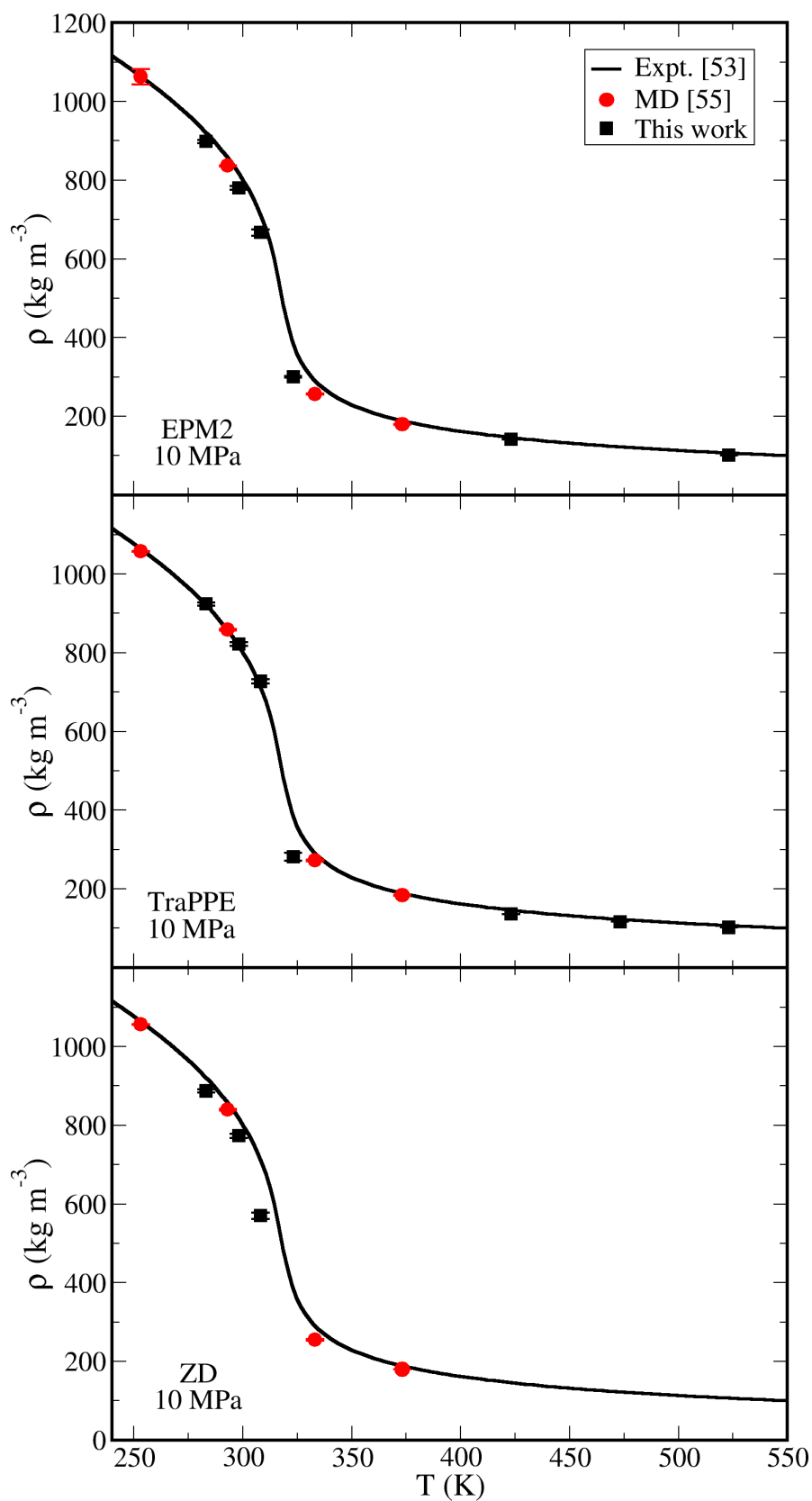


Figure S3. Density of EPM2, TraPPE and ZD CO<sub>2</sub> as a function of temperature at (a) 10 MPa and (b) 100 MPa.

(a)



(b)

