

Note: On the melting points of the TIP4P/2005 and TIP4P/Ice water models using Particle Mesh Ewald for dispersive interactions

— **Supplementary Material** —

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1 Crystallization and melting behaviors per seed for all studied systems

Here we report simulation results for four systems containing 8640 TIP4P/2005 molecules (Table S1), 8640 TIP4P/Ice molecules (Table S2), 4320 TIP4P/2005 molecules (Table S3), and 4320 TIP4P/Ice molecules (Table S4). Figure 1 also shows the potential energy, as a function of time, for the TIP4P/Ice water model with $N = 8640$ molecules at 270.5 and 271.0 K.

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Table S1: Simulation trends for the large system containing $N = 8640$ water molecules, using the TIP4P/2005 water model. Green cells represent scenarios in which the system crystallises partially or completely, whereas red cells represent scenarios in which it melts partially or completely.

Alt text: Table showing crystallization or melting outcomes for each independent seed simulation at temperatures between 248 and 252 K, for the TIP4P/2005 water model with $N = 8640$ molecules.
Alt text: Table showing crystallization or melting outcomes for each independent seed simulation at temperatures between 270 and 274 K, for the TIP4P/Ice water model with $N = 8640$ molecules.

TIP4P/2005 - $N = 8640$ water molecules						
Seed	T (K)					
	248	249	250	250.5	251	252
1	Crystallization	Crystallization	Crystallization	Melting	Melting	Melting
2			Crystallization	Melting	Melting	
3			Crystallization	Melting	Melting	
4			Crystallization	Melting	Melting	
5			Crystallization	Melting	Melting	
6			Crystallization	Melting		
7			Crystallization	Melting		
8			Crystallization	Melting		
9			Crystallization	Melting		
10			Crystallization	Melting		

Table S2: Simulation trends for the large system containing $N = 8640$ water molecules, using the TIP4P/Ice water model. Green cells represent scenarios in which the system crystallises partially or completely, whereas red cells represent scenarios in which it melts partially or completely.

Alt text: Table showing crystallization or melting outcomes for each independent seed simulation at temperatures between 270 and 274 K, for the TIP4P/Ice water model with $N = 8640$ molecules.

TIP4P/Ice - $N = 8640$ water molecules						
Seed	T [K]					
	270	270.5	271	272	273	274
1	Crystallization	Crystallization	Melting	Melting	Melting	Melting
2	Crystallization	Crystallization	Melting	Melting	Melting	
3	Crystallization	Crystallization	Melting	Melting	Melting	
4	Crystallization	Crystallization	Melting			
5	Crystallization	Crystallization	Melting			
6		Crystallization	Melting			
7		Crystallization	Melting			
8		Melting	Melting			
9		Crystallization	Melting			
10		Crystallization	Melting			
11		Crystallization				
12		Crystallization				
13		Crystallization				
14		Crystallization				
15		Melting				
16		Melting				
17		Melting				
18		Crystallization				
19		Crystallization				
20		Crystallization				

Table S3: Simulation trends for the small system containing $N = 4320$ water molecules, using the TIP4P/2005 water model. Green cells represent scenarios in which the system crystallises partially or completely, whereas red cells represent scenarios in which it melts partially or completely.

Alt text: Table showing crystallization or melting outcomes for each independent seed simulation at temperatures between 249 and 252 K, for the TIP4P/2005 water model with $N = 4320$ molecules.

TIP4P/2005 - $N = 4320$ water molecules					
Seed	T [K]				
	249	250	250.5	251	252
1	Crystallization	Crystallization	Crystallization	Melting	Melting
2	Crystallization	Melting	Melting	Melting	Melting
3	Crystallization	Crystallization	Melting	Melting	Melting
4	Crystallization	Crystallization	Melting	Melting	
5	Crystallization	Crystallization	Melting	Melting	
6	Crystallization	Crystallization	Crystallization	Melting	
7	Crystallization	Crystallization	Melting	Melting	
8	Crystallization	Melting	Crystallization	Melting	
9	Crystallization	Crystallization	Melting	Melting	
10	Crystallization	Crystallization	Crystallization	Melting	

Table S4: Simulation trends for the small system containing $N = 4320$ water molecules, using the TIP4P/Ice water model. Green cells represent scenarios in which the system crystallises partially or completely, whereas red cells represent scenarios in which it melts partially or completely.

Alt text: Table showing crystallization or melting outcomes for each independent seed simulation at temperatures between 269 and 272 K, for the TIP4P/Ice water model with $N = 4320$ molecules.

TIP4P/Ice - $N = 4320$ water molecules					
Seed	T [K]				
	269	270	270.5	271	272
1	Crystallization	Crystallization	Crystallization	Melting	Melting
2	Crystallization	Crystallization	Crystallization	Melting	Melting
3	Crystallization	Crystallization	Crystallization	Melting	
4	Crystallization	Crystallization	Crystallization	Melting	
5	Crystallization	Crystallization	Crystallization	Melting	
6	Crystallization	Crystallization	Crystallization	Melting	
7		Crystallization	Melting	Crystallization	
8		Crystallization	Crystallization	Melting	
9		Crystallization	Crystallization	Melting	
10		Crystallization	Melting	Melting	

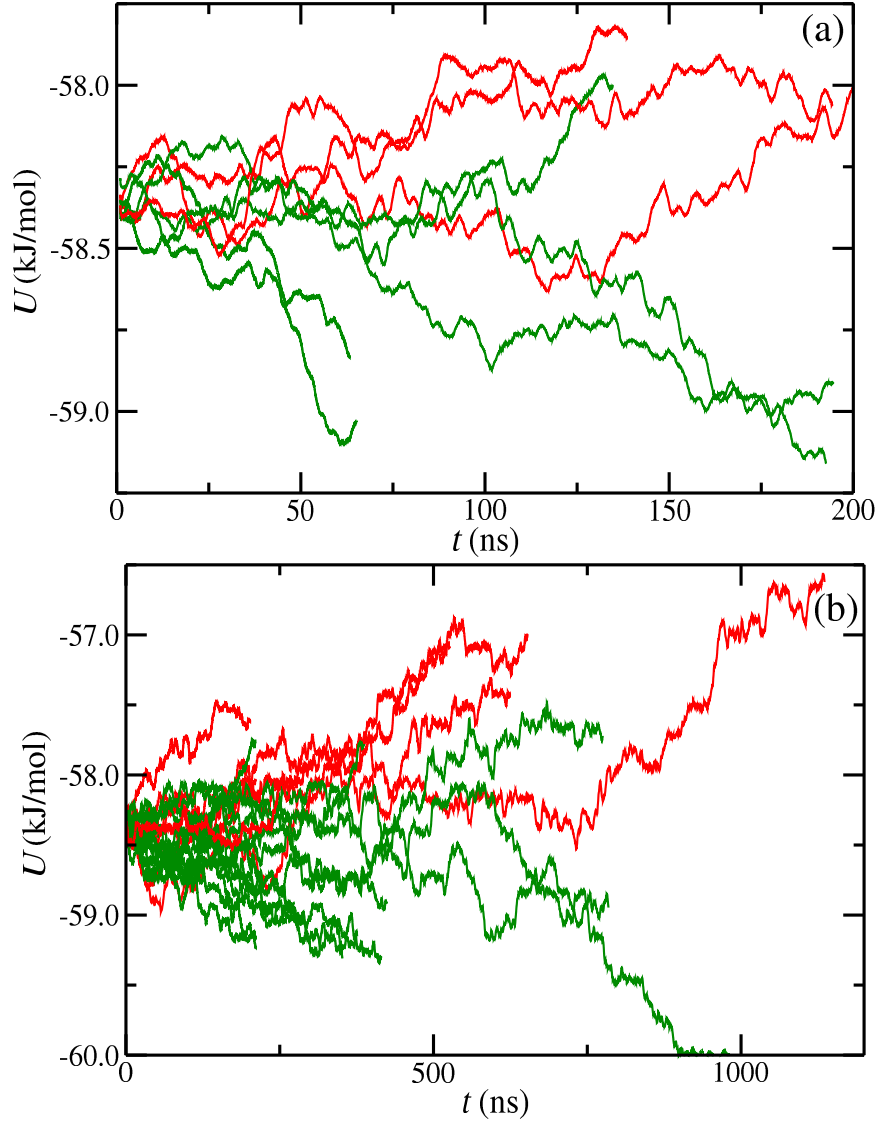


Figure S1: Evolution of the potential energy as a function of time for independent-seed simulations of the TIP4P/Ice water model with run times below 200 ns (a) and above 200 ns (b). Green curves correspond to simulations at 270.5 K (20 seeds), and red curves to simulations performed at 271.0 K (10 seeds). Simulations are performed with $N = 8640$ water molecules. See details in Table S2.

Alt text: Line plot showing the potential energy (in kJ/mol) versus simulation time (in ns) for multiple independent-seed simulations of the TIP4P/Ice water model with 8640 molecules. Panel (a) covers run times below 200 ns and panel (b) above 200 ns. Green curves (20 seeds at 270.5 K) trend downward indicating crystallization, and red curves (10 seeds at 271.0 K) trend upward indicating melting.

2 GROMACS Files

In this section, we describe the molecular dynamics parameter files (.mdp) employed in our simulations, together with the two topology files (.top) used for the TIP4P/Ice and TIP4P/2005 water models.

grompp.mdp

```

1 ; RUN CONTROL PARAMETERS
2 integrator           = md
3 tinit                = 0
4 dt                   = 0.002
5 nsteps               = 500000000
6 init_step            = 0
7 comm-mode            = Linear
8 nstcomm              = 1
9 comm-grps            =
10
11 ; OUTPUT CONTROL OPTIONS
12 nstxout              = 0
13 nstvout              = 0
14 nstfout              = 0
15 nstcheckpoint        = 0
16 nstlog               = 1000
17 nstenergy            = 1000
18 nstxtcout            = 10000
19 xtc-precision        = 10000
20 xtc-grps             =
21 energygrps          =
22
23 ; NEIGHBORSEARCHING PARAMETERS
24 nstlist              = 4
25 ns_type              = grid
26 pbc                  = xyz
27 rlist                = 1.2
28 domain-decomposition = no
29
30 coulombtype          = pme
31 rcoulomb             = 1.2
32 pme_order            = 4
33 fourierspacing       = 0.1
34
35 ; Method for doing Van der Waals
36 vdw-type             = pme
37 rvdw                 = 1.2
38 table-extension      = 1
39
40 ; Temperature coupling
41 Tcoupl               = nose-hoover
42 tau_t                = 2
43 ref_t                = XXX ; Replace with the desired temperature
44 tc-grps              = system
45
46 ; Pressure coupling
47 Pcoupl               = Parrinello-Rahman
48 tau_p                = 2.0
49 pcoupltype           = anisotropic
50 ref_p                = 1 1 1 0 0 0
51 compressibility       = 5e-5 5e-5 5e-5 0 0 0
52
53 ; SIMULATED ANNEALING
54 annealing            = no
55
56 ; GENERATE VELOCITIES FOR STARTUP RUN
57 gen_vel              = yes
58 gen_temp              = XXX ; Replace with the desired temperature
59 gen_seed              = -1
60
61 ; OPTIONS FOR BONDS
62 constraints           = all-angles
63 constraint-algorithm  = Lincs
64 lincs-order          = 4
65 lincs-iter           = 2
66 lincs-warnangle      = 30
67 morse                = no
68 continuation         = no

```

TIP4P/Ice.top

```

1 ;TIP4P/Ice
2
3 [ defaults ]
4 ; nbfunc      comb-rule      gen-pairs      fudgeLJ fudgeQQ
5   1           2             no             1.0      1.0
6
7 [atomtypes]
8 ;name      mass      charge      ptype      sigma      epsilon
9 IW         00.0000      0.00      V       0.00000      0.000000
10 OWT4       15.9994      0.00      A       0.31668      0.882170
11 HW         01.0080      0.00      A       0.00000      0.000000
12
13 [moleculetype]
14 ; name nrexcl
15 water 1
16
17 [atoms]
18 ; nr   type resnr   residu atom cgnr   charge   mass
19 1      OWT4   1      water  OW1   1      0.0000   15.9994
20 2      HW     1      water  HW2   1      0.5897   1.0080
21 3      HW     1      water  HW3   1      0.5897   1.0080
22 4      IW     1      water  MW4   1     -1.1794   0.0000
23
24 [constraints]
25 ;i      j      funct      doh      dhh
26 1        2        1          0.09572
27 1        3        1          0.09572
28 2        3        1          0.15139
29
30 [exclusions]
31 1        2        3        4
32 2        1        3        4
33 3        1        2        4
34 4        1        2        3
35
36 ; The position of the dummy is computed as follows:
37 ;
38 ;           0
39 ;
40 ;           D
41 ;
42 ;           H           H
43 ;
44 ; const = distance (OD) / [ cos (angle(DOH))      * distance (OH) ]
45 ;           0.015 nm      / [ cos (52.26 deg)      * 0.09572 nm      ]
46
47 ; Dummy pos x4 = x1 + a*(x2-x1) + b*(x3-x1)
48
49 ;[dummies3]
50 [ virtual_sites3 ]
51 ; Dummy from      funct      a      b
52 4      1      2      3      1      0.13458      0.13458
53
54 [system]
55 water
56
57 [molecules]
58 ; name nr
59 water N ; Replace with your number of molecules

```

TIP4P/2005.top

```

1 ;TIP4P/2005
2
3 [ defaults ]
4 ; nbfunc      comb-rule      gen-pairs      fudgeLJ fudgeQQ
5   1           2             no             1.0      1.0
6
7 [atomtypes]
8 ;name      mass      charge      ptype      sigma      epsilon
9 IW      00.0000      0.00      V      0.00000      0.000000
10 OWT4     15.9994      0.00      A      0.31589      0.774912
11 HW      01.0080      0.00      A      0.00000      0.000000
12
13 [moleculetype]
14 ; name nrexcl
15 water 1
16
17 [atoms]
18 ; nr  type resnr  residu atom cgnr  charge  mass
19 1    OWT4  1     water  OW1  1     0.0000  15.9994
20 2    HW    1     water  HW2  1     0.5564  1.0080
21 3    HW    1     water  HW3  1     0.5564  1.0080
22 4    IW    1     water  MW4  1    -1.1128  0.0000
23
24 [constraints]
25 ;i      j      funct      doh      dhh
26 1      2      1          0.09572
27 1      3      1          0.09572
28 2      3      1          0.15139
29
30 [exclusions]
31 1      2      3      4
32 2      1      3      4
33 3      1      2      4
34 4      1      2      3
35
36 ; The position of the dummy is computed as follows:
37 ;
38 ;           0
39 ;
40 ;           D
41 ;
42 ;           H           H
43 ;
44 ; const = distance (OD) / [ cos (angle(DOH))      * distance (OH) ]
45 ;           0.015 nm      / [ cos (52.26 deg)      * 0.09572 nm      ]
46
47 ; Dummy pos x4 = x1 + a*(x2-x1) + b*(x3-x1)
48
49 ;[dummies3]
50 [ virtual_sites3 ]
51 ; Dummy from      funct      a      b
52 4      1      2      3      1      0.13193828      0.13193828
53
54 [system]
55 water
56
57 [molecules]
58 ; name nr
59 water N ; Replace with your number of molecules

```