

# Monte Carlo molecular simulation predictions for the heat of vaporization of acetone and butyramide

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## Abstract

Vapor pressure and heats of vaporization are computed for the industrial fluid properties simulation challenge (IFPSC) data set using the Towhee Monte Carlo molecular simulation program. Results are presented for the CHARMM27 and OPLS-aa force fields. Once again, the average result using multiple force fields is a better predictor of the experimental value than either individual force field.

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## 1. Introduction

Our plan was to reproduce an industrial approach to solving the contest problem by taking only 6 weeks to work on the contest using publicly available software and force fields. We predicted the vapor pressures and heats of vaporization without knowledge of the experimental results. Here, we include the simulation results submitted for judging in the contest, along with commentary about the pitfalls encountered during our effort and a discussion of the approach we would suggest for another researcher attempting to compute vapor pressures and heats of vaporization under similar time constraints. It is hoped that this paper enables industrial researchers to assess the effort required to compute these quantities and provides a feeling for the accuracy one should expect from “quick and dirty” molecular simulations.

## 2. Method

The vapor pressures and heats of vaporization for acetone and butyramide were computed using the MCCC'S Towhee simulation package [1]. All simulations were performed in the canonical variant of the Gibbs ensemble [2] at the temperatures specified in the contest information. Simulations were equilibrated for at least 20,000 Monte Carlo cycles (one cycle is  $N$  moves where  $N$  is the number of molecules in the system), and results are reported for simulations of 10,000 cycles. Standard deviations are computed by breaking the simulations into five blocks. Simulations were performed using 120 acetone molecules or 80 butyramide molecules. A butyramide simulation of 10,000 cycles takes roughly 40 h on a single Pentium III 1.4 GHz processor.

The Monte Carlo moves consisted of volume changes, coupled-decoupled configurational bias (CDCB) regrowths, CDCB molecule transfers between the boxes, rotational-bias molecule transfers between the boxes, aggregation volume-bias move type 1 [3] translation of the center-of-mass, and rotation about the center-of-mass. The CDCB algorithm used

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in this work is based on previous work [4,5], but modified to generate trials for the bond lengths and bending angles from a Gaussian distribution centered on the equilibrium value and with a specified standard deviation. This bias was then removed in the acceptance rules. This new method requires an order of magnitude fewer trial sites to achieve the same acceptance rates as the standard method and provides a factor of two speedup in overall simulation time.

The CHARMM27 [6] and OPLS-aa [7] force fields were utilized in this study. The OPLS literature contained all of the parameters needed to simulate acetone and butyramide. However, CHARMM27 was lacking the CT3–CC–CT3 angle parameter for the three carbon atoms in acetone. After consulting with the experts on the charmm.org web site [8], we took the parameters for this angle from the published CT3–CT2–CT2 values. Complete details of the potentials used in this study are shown in the Supplementary information (see Appendix A). A 10 Å cutoff with analytical tail corrections was used to compute the Lennard–Jones interactions, while Coulombic interactions were computed with the Ewald sum method. Charge assignments were inferred from comparison to similar molecules published in the literature.

The heat of vaporization ( $\Delta H_{\text{vap}}$ ) was computed from the average internal energy per molecule, the average vapor pressure, and the average volume per molecule. Starting with the thermodynamic definition of enthalpy ( $H = U + pV$ ),  $\Delta H_{\text{vap}}$  was computed in two different ways. The vapor pressure approach uses

$$\Delta H_{\text{vap}}^{\text{p}} = U_{\text{v}} - U_{\text{l}} + p_{\text{v}} \times (V_{\text{v}} - V_{\text{l}}), \quad (1)$$

where the v and l subscripts refer to the vapor and liquid phases,  $U$  the internal energy per molecule,  $p$  the pressure, and  $V$  is the volume per molecule. This method is satisfying from a theoretical standpoint as it applies the laws of thermodynamics without any assumptions. The pressure calculated in the vapor box is used as a matter of convenience as the observed error bars on liquid box pressures are quite large in a molecular simulation and an equilibrated Gibbs ensemble simulation has the same pressure in both boxes.

The ideal gas law approach for determining  $\Delta H_{\text{vap}}$  again starts with the definition of enthalpy, but then assumes that the liquid phase has a negligible volume per molecule compared

to the vapor phase, and that the vapor phase follows the ideal gas law. The heat of vaporization simplifies to

$$\Delta H_{\text{vap}}^{\text{i}} = U_{\text{v}} - U_{\text{l}} + RT, \quad (2)$$

where  $R$  is the gas constant, and  $T$  is the temperature of the simulation. Note, that one can further assume that the intramolecular energies are the same in both phases, while the intermolecular energies are zero in the vapor phase (allowing this quantity to be computed without any simulation of the vapor phase). The results using such an assumption are discussed below.

### 3. Results and discussion

The simulated and experimental vapor pressures and heats of vaporization are shown in Table 1. Note that the error bars on the low temperature vapor pressures for butyramide are large compared to the magnitude of the vapor pressure due to the low number of accepted molecule transfer moves for these cases (e.g., only 4 in 10,000 cycles for CHARMM27 at 415 K). Additionally, at these low vapor pressures there are less than 10 molecules in the vapor box for all of the butyramide simulations and this also contributes to the poor statistics. We have computed the compressibility ( $pV_{\text{v}}/RT$ ) for each of the vapor phases. If the vapor phase behaved ideally then this ratio would be 1.0. The butyramide results for compressibility are generally quite close to the ideal value of 1.0 regardless of temperature, while the results for acetone show a clear decrease with temperature.

$\Delta H_{\text{vap}}$  results are shown in Table 2. It is curious that the error bars are an order of magnitude larger for the CHARMM27 force field compared with the OPLS force field. In our original submission for the contest (the data shown in Table 2) the error bars for  $\Delta H_{\text{vap}}$  were computed via standard error propagation from the block average error bars of the internal energies (vapor and liquid), the number of molecules in each phase (vapor and liquid), the pressure, and the molar volumes (vapor and liquid). Standard error propagation assumes that the errors are independent, which is a poor assumption for the combination of total internal energy and number of molecules, especially for the intramolecular terms. Thus, while the intramolecular energy per molecule is

Table 1  
Vapor pressures and comparison with ideal gas law

Molecule	$T$ (K)	CHARMM		OPLS		Average Pressure (kPa)	Experimental Pressure (kPa)
		Pressure (kPa)	$pV_{\text{v}}/RT$	Pressure (kPa)	$pV_{\text{v}}/RT$		
Acetone	330	71 <sub>7</sub>	0.98	63 <sub>12</sub>	0.96	67 <sub>7</sub>	104.04 <sub>3</sub>
	375	262 <sub>44</sub>	0.92	295 <sub>42</sub>	0.93	279 <sub>30</sub>	390.3 <sub>10</sub>
	425	946 <sub>153</sub>	0.86	1031 <sub>74</sub>	0.80	989 <sub>85</sub>	1184 <sub>6</sub>
	460	1824 <sub>288</sub>	0.56	1863 <sub>248</sub>	0.68	1844 <sub>190</sub>	2225 <sub>11</sub>
Butyramide	415	5 <sub>1</sub>	0.94	1.9 <sub>1</sub>	1.00	3.5 <sub>5</sub>	4.65 <sub>5</sub>
	455	36 <sub>7</sub>	0.98	15 <sub>2</sub>	0.97	25 <sub>3</sub>	22.3 <sub>2</sub>
	490	77 <sub>21</sub>	1.01	41 <sub>13</sub>	1.02	59 <sub>12</sub>	67.7 <sub>7</sub>
	520	212 <sub>23</sub>	0.89	83 <sub>9</sub>	1.00	148 <sub>12</sub>	151.6 <sub>3</sub>

The statistical error in the last digits is shown as a subscript for the vapor pressures.

Table 2  
Heat of vaporization computed using the vapor pressure (Eq. (1)) and the ideal gas law (Eq. (2))

Molecule	$T$ (K)	CHARMM		OPLS		Average		Experimental
		$\Delta H_{\text{vap}}^{\text{p}}$ (kJ/mol)	$\Delta H_{\text{vap}}^{\text{i}}$ (kJ/mol)	$\Delta H_{\text{vap}}^{\text{p}}$ (kJ/mol)	$\Delta H_{\text{vap}}^{\text{i}}$ (kJ/mol)	$\Delta H_{\text{vap}}^{\text{p}}$ (kJ/mol)	$\Delta H_{\text{vap}}^{\text{i}}$ (kJ/mol)	
Acetone	330	33 <sub>11</sub>	33 <sub>11</sub>	34 <sub>3</sub>	34 <sub>2</sub>	33 <sub>6</sub>	33 <sub>6</sub>	29.07 <sub>15</sub>
	375	30 <sub>23</sub>	30 <sub>23</sub>	31 <sub>2</sub>	31 <sub>2</sub>	31 <sub>12</sub>	31 <sub>12</sub>	25.92 <sub>13</sub>
	425	25 <sub>23</sub>	26 <sub>23</sub>	26 <sub>2</sub>	27 <sub>2</sub>	26 <sub>11</sub>	27 <sub>11</sub>	21.4 <sub>4</sub>
	460	20 <sub>30</sub>	22 <sub>30</sub>	22 <sub>2</sub>	24 <sub>1</sub>	21 <sub>15</sub>	23 <sub>15</sub>	17.1 <sub>4</sub>
Butyramide	415	49 <sub>11</sub>	50 <sub>11</sub>	69 <sub>2</sub>	69 <sub>2</sub>	59 <sub>6</sub>	59 <sub>6</sub>	63.0 <sub>16</sub>
	455	62 <sub>30</sub>	63 <sub>30</sub>	62 <sub>5</sub>	62 <sub>5</sub>	63 <sub>15</sub>	63 <sub>15</sub>	59.3 <sub>12</sub>
	490	46 <sub>29</sub>	46 <sub>28</sub>	61 <sub>7</sub>	61 <sub>6</sub>	53 <sub>15</sub>	53 <sub>15</sub>	56.2 <sub>12</sub>
	520	42 <sub>10</sub>	42 <sub>10</sub>	58 <sub>3</sub>	58 <sub>3</sub>	50 <sub>5</sub>	50 <sub>5</sub>	53.4 <sub>2</sub>

The subscripts show the statistical error in the final digits.

Table 3  
Average intermolecular and intramolecular energies (kJ/mol) per molecule for acetone at 30 K

Force field	$U^{\text{inter}}$			$U^{\text{intra}}$		
	Liquid	Vapor	$\Delta U_{\text{vap}}^{\text{inter}}$	Liquid	Vapor	$\Delta U_{\text{vap}}^{\text{intra}}$
CHARMM	−0.6	29.6	30.2	−178.07	−178.02	0.05
OPLS	−18.5	12.97	31.5	−22.9	−23.2	−0.4

nearly constant, it contributes to our estimated error bars due to the fluctuations in the number of molecules in each simulation box. The substantially larger intramolecular energies for the CHARMM27 force field, shown in Table 3, result in larger error estimates. We have subsequently performed additional simulations for acetone at 330 K computing the error bars using the block average of  $\Delta H_{\text{vap}}^{\text{v}}$  resulting in  $32.6 \pm 0.4$  kJ/mol for CHARMM27 and  $33.5 \pm 0.2$  kJ/mol for OPLS. These results agree with our original values and have error bars that are better in line with the observed reproducibility of the acetone enthalpy results.

Table 3 shows that the CHARMM27 and OPLS force fields have very different liquid and vapor phase internal energies for acetone despite the fact that the difference in the vapor and liquid energies ( $\Delta U_{\text{vap}}$ ) is very similar. These results illustrate that computing the heat of vaporization from a single, liquid box simulation introduces large errors. The intermolecular energies in the vapor box are significant contributors to the internal energy, especially when the molecule includes Coulombic interactions, and therefore cannot be neglected.

The butyramide simulations show unusual behavior for the heats of vaporization as they do not follow the expected trend of decreasing monotonically with increasing temperature. This indicates poor sampling due to the low acceptance rate for molecule transfer moves. Strangely, for the CHARMM27 butyramide simulations the rotational-bias molecule transfer move had a substantially higher acceptance rate than the configurational-bias molecule transfer move. It appears that the high intramolecular energies are causing trouble for the configurational-bias algorithm in this case and suggests CHARMM27 butyramide as a challenging test case for future work on conformation sampling algorithms.

There is little difference between the heats of vaporization predicted using the vapor pressure or those predicted using

the ideal gas law. This occurs because the internal energy of vaporization is the dominant term in our simulations, and also because these simulations are performed in a region of the vapor–liquid coexistence curve where the ideal gas law is a good approximation. However, as the compressibility begins to deviate substantially from unity (acetone at 460 K) the heats of vaporization become noticeably different. We suggest using the vapor pressure method for future work as it is formally more correct than the ideal gas approximation and does not require more simulation effort.

#### 4. Conclusions

With the publication of the experimental results [9], we can assess the accuracy of these force fields for predicting vapor pressure and heats of vaporization. For acetone, CHARMM27 and OPLS gave results in good agreement with each other, but both under-predicted the vapor pressures by roughly 30% and over-predicted the heats of vaporization by roughly 3 kJ/mol. In contrast, the butyramide CHARMM27 pressures are a factor of two larger than those predicted by OPLS, yet the average result submitted for the contest fortuitously agrees very well with the experimental data. The heats of vaporization for CHARMM27 butyramide are clearly not equilibrated sufficiently as they do not show the near-universal trend of decreasing heat of vaporization with increasing temperature. In particular, the 455 K data point is wildly out of range compared with the other temperatures for CHARMM27. The average results agree fairly well with the experimental data as the force fields deviate from experiment in opposite directions. As was observed in our entry for the first IFPSC [4], the average values from multiple force fields were once again in better agreement with experiment than any of the individual force fields. We are currently working

on a larger scale study to see if this is a general trend, or just a lucky anomaly.

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## Appendix A. Supplementary information

See Tables A.1–A.6.

The CHARMM27 force field uses the following equations to represent the internal energy.

$$U_{\text{nonbond}}(r_{ij}) = 4\epsilon_{ij} \left[ \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left( \frac{\sigma_{ij}}{r_{ij}} \right)^6 \right] + \frac{q_i q_j}{r_{ij}}$$

$$\epsilon_{ij} = \sqrt{\epsilon_{ii}\epsilon_{jj}}$$

$$\sigma_{ij} = 0.5(\sigma_{ii} + \sigma_{jj})$$

$$U_{\text{bond}}(r_{ij}) = k_1[r_{ij} - k_0]^2$$

$$U_{\text{angle}}(r_{ik}, \theta_{ijk}) = k_1[\theta_{ijk} - \theta_0]^2 + k_3[r_{ik} - k_2]^2$$

$$U_{\text{dihed}}(r_{il}, \phi_{ijkl}) = k_1[1 + \cos(k_2\phi_{ijkl} - k_3)]$$

$$+ 4\epsilon_{il}^{1-4} \left[ \left( \frac{\sigma_{il}^{1-4}}{r_{il}} \right)^{12} - \left( \frac{\sigma_{il}^{1-4}}{r_{il}} \right)^6 \right] + \frac{q_i q_l}{r_{il}}$$

Table A.1

Nonbonded Towhee atom type names and parameters for CHARMM27

Atom	Type	$\epsilon$	$\sigma$	Charge	$\epsilon^{1-4}$	$\sigma^{1-4}$
Acetone						
C(H <sub>3</sub> )	CT3	40.257	3.671	-0.27	5.032	3.385
C(=O)	CC	35.225	3.564	0.55	35.225	3.564
O(=C)	O	60.386	3.029	-0.55	60.386	2.495
H	HA	11.071	2.352	0.09	11.071	2.352
Butyramide						
C(H <sub>3</sub> )	CT3	40.257	3.671	-0.27	5.032	3.385
C(H <sub>2</sub> )	CT2	27.677	3.875	-0.18	5.032	3.385
C(=O)	CC	35.225	3.564	0.55	35.225	3.564
N	NH <sub>2</sub>	100.643	3.296	-0.62	100.643	3.296
H(C)	HA	11.071	2.352	0.09	11.071	2.352
O	O	60.386	3.029	-0.55	60.386	2.495
H(N)	H	23.148	0.400	0.31	23.148	0.400

Energy units are in K and distances are in Å.

Table A.2

Bond and angle atom type names and parameters for CHARMM27 acetone and butyramide

Bond	$k_0$	$k_1$		
CC-CT2	1.522	100643.3		
CC-CT3	1.522	100643.3		
C-NH <sub>2</sub>	1.360	216383.1		
C-O	1.230	327090.8		
CT2-CT2	1.530	111965.7		
CT2-CT3	1.528	111965.7		
CT2-HA	1.111	155493.9		
CT3-HA	1.111	162035.8		
H-NH <sub>2</sub>	1.000	241544.0		
Angle	$\theta_0$	$k_1$	$k_2$	$k_3$
CT2-CT2-CC	108.000	26167.3	0.0	0.0
CT2-CT2-CT3	115.000	29186.6	2.5610	4025.7
CT3-CC-CT3	115.000	29186.6	2.5610	4025.7
CT2-CT2-HA	110.100	13335.2	2.1790	11337.5
HA-CT2-HA	109.000	17864.2	1.8020	2717.4
CT2-CT3-HA	110.100	17411.3	2.1790	11337.5
CT3-CT2-HA	110.100	17411.3	2.1790	11337.5
CC-CT2-HA	109.500	16606.1	2.1630	15096.5
CC-CT3-HA	109.500	16606.1	2.1630	15096.5
HA-CT3-HA	108.400	17864.2	1.8020	2717.4
CC-NH <sub>2</sub> -H	120.000	25160.8	0.0	0.0
H-NH <sub>2</sub> -H	120.000	11574.0	0.0	0.0
CT2-CC-NH <sub>2</sub>	116.500	25160.8	2.4500	25160.8
CT2-CC-O	121.000	7548.2	2.4400	25160.8
CT3-CC-O	121.000	7548.2	2.4400	25160.8
NH <sub>2</sub> -CC-O	122.500	37741.2	2.3700	25160.8

Energy units are in K, distances are in Å, and angles are in degrees.

Table A.3

Torsion atom type names and parameters for CHARMM27 acetone and butyramide

Dihedral	$k_1$	$k_2$	$k_3$
HA-CT2-CC-NH <sub>2</sub>	0.0	0.0	0.0
HA-CT2-CC-O	0.0	0.0	0.0
x-CT2-CT3-x	80.5	3	0.0
x-CT2-CT2-x	25.2	6	$\pi$
x-CC-CT3-x	25.2	6	$\pi$
CT2-CC-CT2-NH <sub>2</sub>	25.2	6	$\pi$
O-CC-CT2-CT2	25.2	6	$\pi$
x-CC-NH <sub>2</sub> -x	704.5	2	$\pi$
x-CT2-CT2-x	98.1	3	0.0
Improper	$k_0$	$k_1$	
NH <sub>2</sub> : CC,H,H	2012.9	0.0	
CC: CT2,NH <sub>2</sub> ,O	22644.7	0.0	
CC: NH <sub>2</sub> ,CT2,O	22644.7	0.0	

Energy units are in K, distances are in Å, and angles are in radians.

$$U_{\text{improper}}(\phi_{ijkl}) = k_0[\phi_{ijkl} - k_1]^2$$

The OPLS force field uses the following equations to represent the internal energy.

$$U_{\text{nonbond}}(r_{ij}) = 4\epsilon_{ij} \left[ \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left( \frac{\sigma_{ij}}{r_{ij}} \right)^6 \right] + \frac{q_i q_j}{r_{ij}}$$

$$\epsilon_{ij} = \sqrt{\epsilon_{ii}\epsilon_{jj}}$$

Table A.4  
Nonbonded Towhee atom type names and parameters for the OPLS force field

Atom	Type	$\epsilon$	$\sigma$	Charge
Acetone				
C(H <sub>3</sub> )	CT	33.212	3.500	-0.18
C(=O)	C k	52.838	3.750	0.47
O(=C)	O	105.675	2.960	-0.47
H	HC~1	15.096	2.500	0.06
Butyramide				
C(H <sub>3</sub> )	CT	33.212	3.500	-0.18
C(H <sub>2</sub> )	CT	33.212	3.500	-0.12
C(=O)	C n	52.838	3.750	0.50
N	N	85.547	3.250	-0.76
H(C)	HC~1	15.096	2.500	0.06
O	O	105.675	2.960	-0.50
H(N)	H	0.0	0.0	0.38

Energy units are in K and distances are in Å.

Table A.5  
Bond and angle Towhee atom type names and parameters for OPLS acetone and butyramide

Bond	$k_0$	$k_1$
C k-CT	1.522	159519.7
C n-CT	1.522	159519.7
C n-N	1.335	246576.1
C k-O	1.229	286833.5
C n-O	1.229	286833.5
CT-CT	1.529	134862.1
CT-HA	1.09	171093.6
H-N	1.010	218396.0
Angle	$\theta_0$	$k_1$
H-N-H	120.000	17612.6
C n-CT-CT	111.100	31702.6
CT-C n-N	116.600	35225.2
CT-C k-O	120.400	40257.3
CT-C n-O	120.400	40257.3
C k-CT3-HC~1	109.500	17612.6
C n-CT3-HC~1	109.500	17612.6
N-C n-O	122.900	40257.3
CT-C k-CT	116.000	35225.2
HC~1-CT3-HC~1	107.800	16606.1
CT-CT-HC~1	110.700	18870.6
CT-CT-CT	112.700	29362.7
C n-N-H	119.800	17612.6

Energy units are in K, distances are in Å, and angles are in degrees.

Table A.6  
Torsion Towhee atom type names and parameters for OPLS acetone and butyramide

Dihedral	$k_1$	$k_2$	$k_3$
C n-CT-CT-H~1	0.0	0.0	-25.2
C n-CT-CT-CT	-518.3	-78.8	79.3
CT-C k-CT-HC~1	0.0	0.0	69.2
CT-C n-N-H	0.0	1232.9	0.0
CT-CT-C n-N	817.7	-101.1	-34.2
CT-CT-C n-O	0.0	293.4	0.0
CT-CT-CT-HC~1	0.0	0.0	92.1
HC~1-CT-C k-O	0.0	0.0	0.0
HC~1-CT-C n-N	0.0	0.0	0.0
HC~1-CT-CT-HC~1	0.0	0.0	80.0
Improper	$k_1$	$k_2$	$k_3$
C n: O,CT,N	0.0	5283.8	0.0
N: C n,H,H	0.0	503.2	0.0

Energy units are in K and distances are in Å.

$$\sigma_{ij} = \sqrt{\sigma_{ii}\sigma_{jj}}$$

$$U_{\text{bond}}(r_{ij}) = k_1[r_{ij} - k_0]^2$$

$$U_{\text{angle}}(\theta_{ijk}) = k_1[\theta_{ijk} - \theta_0]^2$$

$$U_{\text{dihed}}(r_{il}, \phi_{ijkl}) = k_1[1 + \cos(\phi_{ijkl})] + k_2[1 - \cos(2\phi_{ijkl})] + k_3[1 + \cos(3\phi_{ijkl})] + \frac{U_{\text{nonbond}}(r_{il})}{2}$$

$$U_{\text{improper}}(\phi_{ijkl}) = k_1[1 + \cos(\phi_{ijkl})] + k_2[1 - \cos(2\phi_{ijkl})] + k_3[1 + \cos(3\phi_{ijkl})]$$

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