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Molecular Dynamics Simulations of Liquid Nitromethane

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A potential energy function with harmonic intramolecular and Lennard-Jones plus Coulombic intermolecular terms was tested in molecular dynamics simulations of liquid nitromethane. Parameter values were adjusted iteratively until satisfactory agreement with density functional pair calculations and experimental data was achieved. The properties computed using the *NVT* and *NPT* ensembles were the heat of vaporization, dielectric constant, self-diffusion coefficient, density, heat capacity at constant pressure, pair correlation functions, single molecule and collective dipole moment reorientation times, the vibrational spectrum, and the effect of increasing pressure upon the C–N stretching frequency. Overall, the results were in reasonable accord with experimental results, the greatest discrepancy being for the dielectric constant. It was concluded, on the basis of the reorientation times and the calculated molecular surface electrostatic potential, that the intermolecular interactions in liquid nitromethane at 1 atm are not highly directional and site-specific.

Introduction

Molecular dynamics simulations have come to be an important tool for studying the initiation and propagation of detonation in energetic compounds. (For a recent brief review, see ref 1.) The general objectives are to achieve a better understanding of the process and the factors that are involved, e.g., the localization and transfer of energy, vibrational excitation and bond-breaking, lattice disruption and the role of defects, etc. An eventual goal is to minimize vulnerability to unintended detonations caused by accidental external stimuli, such as impact and shock.

Some of the simulations have dealt with idealized model systems; for example, the lattice might be monatomic^{2–4} or two-dimensional.^{2,3,5–8} In studies of nitromethane, it has sometimes been represented simply by the diatomic CN.^{5,9–11} There have also been efforts to simulate actual energetic compounds. In principle, this requires appropriate potential functions for describing both the inter- and intramolecular interactions in the system. A possible first step, however, is to treat just an isolated molecule, thereby eliminating the need for intermolecular potentials. This has been done for several nitramines.^{12–15} Another option is to build the crystal lattice and to view it as composed of rigid molecules (i.e., fixed structures). Then only intermolecular interactions need be considered. Such a procedure has now been applied to a large number of energetic compounds,^{16,17} using a potential function that was shown to be transferable between different chemical classes.

The next step, and our objective in the present work, is to develop an approach that takes account of both inter- and intramolecular interactions in order that the simulations can eventually include such features as vibrational excitation, molecular rearrangement, and bond breaking. (These are excluded by the rigid molecule assumption.) Our initial focus is on nitromethane. While it is a liquid at room temperature and pressure, a satisfactory treatment of it should be applicable as well to solids. (A study with a similar objective has very recently been carried out for liquid dimethylnitramine.¹⁸)

Methods

A. Potential Function. All simulations were performed with the molecular dynamics code CHARMM,^{19,20} version c25b1, using the potential energy function,

$$V = \sum_{\text{bonds}} K_R (R - R_0)^2 + \sum_{\text{bond angles}} K_\theta (\theta - \theta_0)^2 + \sum_{i \text{ on A}} \sum_{j \text{ on B}} \left\{ (\epsilon_i \epsilon_j)^{1/2} \left[\left(\frac{0.5(\sigma_i + \sigma_j)}{r_{ij}} \right)^{12} - \left(\frac{0.5(\sigma_i + \sigma_j)}{r_{ij}} \right)^6 \right] + \frac{q_i q_j}{\epsilon r_{ij}} \right\} \quad (1)$$

The first two summations in eq 1 describe intramolecular interactions between bonded atoms; the quantities $(R - R_0)$ and $(\theta - \theta_0)$ are the displacements from the equilibrium bond lengths and angles. The third summation encompasses Lennard-Jones and Coulombic interactions and would involve (for nitromethane) each pair of atoms i and j on different molecules A and B separated by r_{ij} and having charges q_i and q_j . (For larger molecules, some intramolecular Lennard-Jones and Coulombic interactions are also taken into account.^{19,20}) We did not include Urey–Bradley, torsional, or out-of-plane angular terms²⁰ in eq 1 because (a) the rotational barrier of nitromethane is nearly zero²¹ and (b) the vibrational spectrum of nitromethane was found to be reproduced satisfactorily without their inclusion. The assignment of the parameters K_R , K_θ , ϵ_i , ϵ_j , σ_i and σ_j , as well as the atomic charges, will be discussed below. ϵ was set equal to 1.

Since the bond-related terms in eq 1 are harmonic, dissociation cannot be simulated directly. It can be inferred, however, since a bond that would in reality undergo dissociation would in the present simulations be observed to have considerably increased vibrational energy and large-amplitude oscillations.¹⁵

B. Parametrization. The starting nitromethane molecular geometry was optimized by a B3P86/6-31+G* density func-

TABLE 1: Final Input Data for Intramolecular Interactions

bond	R_0 (Å)	K_R (kcal mol ⁻¹ Å ⁻²)
N—O	1.222	680.0
C—N	1.492	335.0
H—C	1.086	360.0

angle	θ_0 (deg)	K_θ (kcal mol ⁻¹ rad ⁻²)
O—N—O	125.6	60.0
C—N—O	117.9	60.0
H—C—N	107.6	51.5
H—C—H	111.3	40.0

TABLE 2: Final Input Data for Intermolecular Interactions

atom	q (au)	ϵ (kcal/mol)	σ (Å)
H	+0.142	0.0220	1.306
C	-0.331	0.0800	2.138
N	+0.717	0.2000	1.999
O	-0.406	0.1200	1.683

tional calculation, using the Gaussian 94 code²² in conjunction with the Becke three-parameter hybrid²³ and Perdew-86 functionals.²⁴ The initial parameter values for eq 1 were taken from the CHARMM protein library, and the atomic charges were from a previous nitromethane calculation.²⁵ An iterative parametrization process was subsequently carried out, which focused alternately on the intermolecular and intramolecular interactions. First, successive simulations were performed of nitromethane in the liquid state (as it is at 300 K), with the Lennard-Jones parameters being adjusted each time until the computed intermolecular potential energy was acceptably close in magnitude to the observed heat of vaporization (9.147 kcal/mol at 298 K²⁶) and the pair correlation functions $g(r)$ were similar to those obtained earlier.²⁵ Next, the stretching and bending force constants, K_R and K_θ , were varied until the vibrational frequencies of an isolated molecule reproduced reasonably well the experimental gas-phase spectrum²⁷ and the normal modes were, as much as possible, orthogonal (i.e., we sought to minimize the energy contributions to a given mode from the others). At this point, additional simulations were carried out to check if the $g(r)$ and the heat of vaporization were still satisfactory and to make any necessary modifications in the Lennard-Jones parameters. Finally, the strongest pair interaction energy computed with CHARMM was compared to that resulting from density functional B3P86/6-31+G** calculations and the Lennard-Jones parameters were scaled so as to achieve good agreement. The entire iterative process was repeated as needed to obtain an overall satisfactory fit to the database. The atomic charges and the final parameter values are listed in Tables 1 and 2.

C. Simulation Methodology. All simulations were for liquid nitromethane. The system consisted of 216 molecules in a cubic cell of side 26.8 Å, corresponding to an initial density of 1.137 g/cm³.²⁶ One long simulation was performed in the *NVT* ensemble (constant number of molecules, volume, and temperature); the others were at constant pressure (*NPT*).

Periodic boundary conditions were enforced by creating images of the atoms in the primary simulation cell, which track the motion of the latter (via continual coordinate transformations) and replace them in the cell if they move out of it. Intermolecular interactions were handled on a group basis (e.g., CH₃, NO₂) and included both primary–primary, i – j , and primary–image, i – j' , contributions. Temperature and pressure control was handled by the Berendsen thermostat/barostat.²⁸ The coupling constants τ_T and τ_P were assigned values such as to

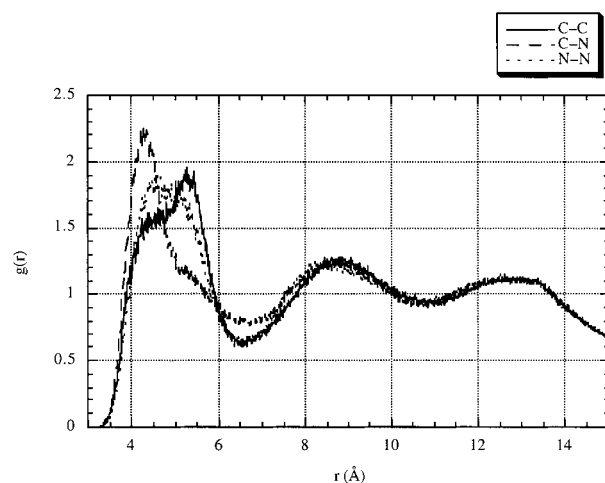


Figure 1. C–C, C–N, and N–N pair correlation functions for nitromethane.

maintain constant energy fluctuations and adequately restrict temperature and pressure variations, as described previously.²⁹ They were initially obtained at 300 K and 1 atm and reevaluated if necessary at other temperatures and pressures.

The time step was 0.5 fs in all simulations, and the nonbonded neighbor list was updated every 20 steps, using a cutoff of 14 Å. Lennard-Jones interactions were truncated at 12 Å; switching was employed between 10 and 12 Å. Coulombic interactions were calculated explicitly to 12 Å, and beyond that by the Ewald particle mesh method,³⁰ in order to properly account for long-range electrostatic interactions. The dielectric constant is extremely sensitive to these.^{31,32} The Verlet integration algorithm was employed.³³

A simulation of 500 ps duration was performed in the *NVT* ensemble at 300 K to determine the properties of the model under conventional liquid-state simulation conditions. The exceptional length of this run was required because the dielectric constant, which was being evaluated, is known to converge particularly slowly.³⁴ Other properties calculated were the pair correlation functions, the self-diffusion coefficient D , the vibrational frequencies and the energy contributions to the normal modes, and the single molecule and collective dipole moment reorientation times (τ_S and τ_C). *NPT* simulations of 50 or 100 ps were carried out to obtain the heat of vaporization ΔH_{vap} , the density, the heat capacity at constant pressure C_P , and the effect of increasing pressure on the C–N stretching frequency.

Results

A. Pair Correlation Functions. Figure 1 shows the C–C, C–N, and N–N pair correlation functions; the N–O and O–O are in Figure 2. Thus, for a given nitromethane molecule, the nearest-neighbor shell is at 4–6 Å, the next is at roughly 9 Å, and there are indications of a third at approximately 13 Å.

The pair correlation functions for an oxygen on one molecule and the three hydrogens on another are in Figure 3. There is relatively little structure, and no distinction between the hydrogens is evident. While the possibility of C–H...O hydrogen bonding is now accepted,^{35,36} it seems unlikely that the two peaks in the 2–3 Å region represent such interactions, since these distances also correspond to intramolecular O...H separations in nitromethane. This will be further discussed below. The small peak at about 4.5 Å is too far away to be indicative of hydrogen bonding.

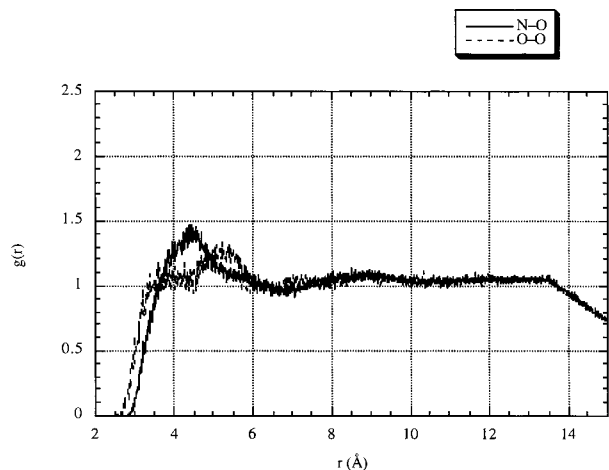


Figure 2. N–O and O–O pair correlation functions for nitromethane.

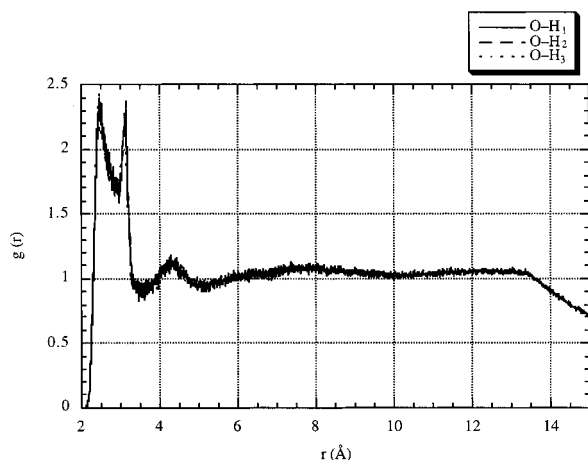


Figure 3. Pair correlation functions for an oxygen with each of the three hydrogens.

TABLE 3: Calculated and Experimental Liquid Properties

property	simulation result	experimental
ΔH_{vap}	9.50 kcal mol ⁻¹ (300 K) 8.20 kcal mol ⁻¹ (400 K)	9.147 kcal mol ⁻¹ (298.15 K) ^a 8.124 kcal mol ⁻¹ (374.4 K) ^b
ϵ_0	26.44	35.87 ^c
density	1.107 g cm ⁻³	1.137 g cm ⁻³ ^d
C_p	113.6 J K ⁻¹ mol ⁻¹	105.98 J K ⁻¹ mol ⁻¹ ^a
D	1.52×10^{-5} cm ² s ⁻¹	

^a Reference 37. ^b Reference 38. ^c Reference 40. ^d Reference 26.

B. Liquid Properties (Table 3). The predicted heat of vaporization ΔH_{vap} (the negative of the calculated intermolecular potential energy) is 9.50 kcal/mol at 300 K and 8.20 kcal/mol at 400 K; these are in very good agreement with the observed 9.147 kcal/mol at 298.15 K³⁷ and 8.124 kcal/mol at 374.4 K.³⁸

The dielectric constant ϵ_0 was obtained from the G_k factor,³⁹

$$G_k = \frac{\langle \vec{M}^2 \rangle}{N \langle \vec{\mu}^2 \rangle} \quad (2)$$

in which

$$\vec{M} = \sum_A \vec{\mu}_A \quad (3)$$

with $\vec{\mu}_A$ being the dipole moment of molecule A and N the total number of molecules. G_k is related to ϵ_0 by

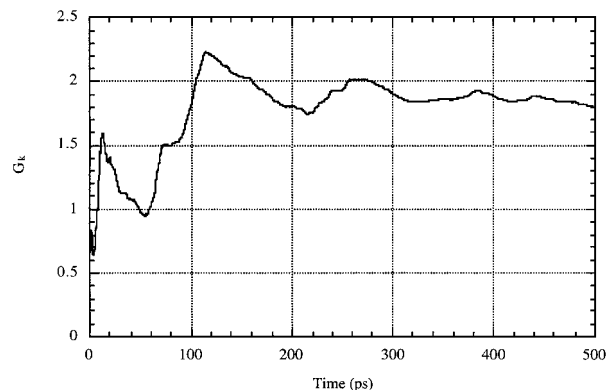


Figure 4. Time dependence of cumulative G_k factor.

$$G_k = \frac{3TV(\epsilon_0 - 1)}{4\pi N \langle \vec{\mu}^2 \rangle} \quad (4)$$

As seen in Figure 4, G_k fluctuates considerably for nearly 350 ps, after which it begins to converge. This demonstrates the need for long simulations in calculating this property, as mentioned above. The resulting ϵ_0 , 26.44, is a fair approximation to the experimental 35.87.⁴⁰

The self-diffusion coefficient D was estimated from the time dependence of the molecular mean-square displacements.⁴¹ Our prediction, $D = 1.52 \times 10^{-5}$ cm²/s, is quite reasonable in comparison to measured values for other systems (e.g., water, ethanol, etc.);^{26,39} however, we are not aware of one for nitromethane.

In an *NPT* simulation at 300 K and 1 atm, the volume was found to change only slightly. Accordingly, the final calculated density, 1.107 g/cm³, was very close to the initial (experimental) value, 1.137 g/cm³.²⁶

The heat capacity at constant pressure, C_p , was determined from its definition and a finite differences approximation,²⁹ using data obtained at 290 and 300 K and 1 atm pressure. The result, 113.6 J K⁻¹ mol⁻¹, agrees well with the experimental 105.98 J K⁻¹ mol⁻¹.³⁷

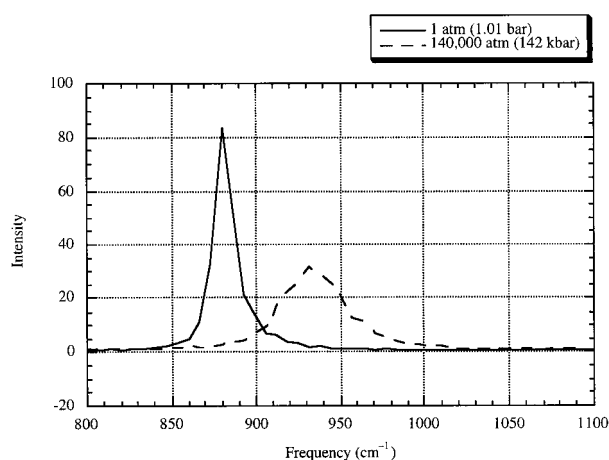
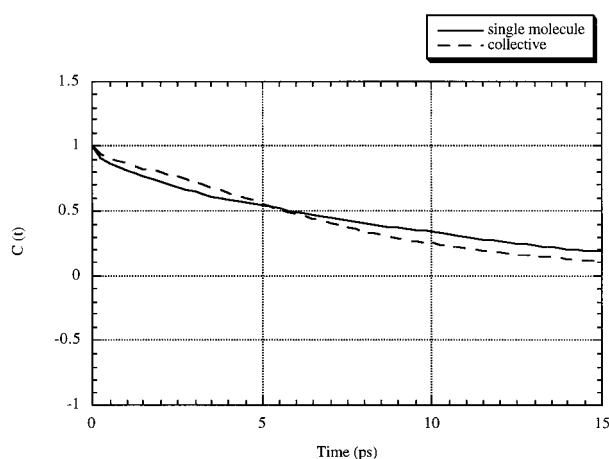
C. Vibrational Frequencies. The calculated and observed⁴² vibrational frequencies of liquid nitromethane are compared in Table 4. The agreement is overall satisfactory, although in four instances the difference is 100 cm⁻¹ or more. The absence of anharmonicity in the intramolecular force field is presumably one source of error. In addition, some of the discrepancies reflect the fact that there is known to be significant mixing among some of the modes.^{27,42} Evidence of this in the present work is that four of the modes listed in Table 4 have energetic contributions of at least 30% from others. The three largest differences between calculated and observed frequencies in Table 4 are for modes in which mixing has already been noted in other studies.^{27,42}

Since one of our eventual objectives is to simulate the propagation of shock waves through energetic crystal lattices, we investigated the ability of our model to reproduce the effect of very high pressure on, specifically, the C–N stretching frequency. (Attention has been focused on the C–N bond as possibly playing an important role in the decomposition of nitromethane.^{43–45}) Figure 5 shows the C–N frequencies computed in the *NPT* ensemble at 1 atm (1.01 bar) and at 140 000 atm (142 kbar). The predicted change of +52 cm⁻¹ is in excellent agreement with that reported by Pangilinan and Gupta.⁴⁵ (They, as well as Moore and Schmidt,⁴⁶ also discuss possible complications due to hot bands.)

TABLE 4: Vibrational Frequencies and Energetic Contributions to Normal Modes^a

mode	ν (calcd), cm ⁻¹	ν (exptl), cm ⁻¹	energy contributions, %
r(NO ₂)	451	480	93
δ_s (NO ₂)	518	655	85
ν (CN)	883	917	ν (CN), 52; ν_s (NO ₂), 37
r(CH ₃)	1073	1103	85
r'(CH ₃)	1092	1125	88
ν_s (NO ₂)	1349	1402	ν_s (NO ₂), 48; δ_s (CH ₃), 38
δ_a (CH ₃)	1521	1426	87
δ_a' (CH ₃)	1526	1426	69
δ_s (CH ₃)	1548	1379	δ_s (CH ₃), 45; δ_a' (CH ₃), 19; ν (CN), 18; ν_s (NO ₂), 11
ν_a (NO ₂)	1735	1561	90
ν_s (CH ₃)	2941	2955	100
ν_a (CH ₃)	3075	3050	90
ν_a' (CH ₃)	3076	3045	90

^a Experimental frequencies are from ref 42. If only one energy contribution is given, it is from the mode itself.

**Figure 5.** C–N stretching vibration at two different pressures.**Figure 6.** Autocorrelation functions for single molecule and collective dipole moments.

Discussion

It is widely accepted that there are significant intermolecular interactions in liquid nitromethane, particularly at higher pressures.^{25,27,42,45–49} To better characterize them, our NVT simulation included a determination of the autocorrelation functions for a single molecule and collective dipole moments.³⁹ These were found to be very similar (Figure 6), as are accordingly the corresponding reorientation (or relaxation) times τ_s and τ_c , estimated (from the slopes of $\ln C$ vs t) to be 9 and

7 ps, respectively. This indicates that the intermolecular interactions are not strongly directional, since that would cause the collective decay rate to be considerably lower than the single molecule, and $\tau_c > \tau_s$. In liquid water, for example, the τ_c/τ_s ratio at 300 K is 3.7 (based on experimental values³⁹). Our results are fully consistent with those of Giorgini et al.,⁵⁰ who obtained reorientation times through Raman and Rayleigh light-scattering studies and concluded that “static orientational correlation effects are insignificant in liquid nitromethane” and that the orientational dynamics are those of the single molecule. Thus, whatever is the nature of the intermolecular interactions, they do not produce, at 1 atm, a collective structure with a substantial degree of persistent order.

Analyses of crystal structures⁵¹ and a subsequent computational study⁵² have drawn attention to attractive intermolecular N...O interactions that can occur between nitro groups. However, Figure 2 indicates that these are not likely to be important in liquid nitromethane, since the first N...O peak is at about 4.5 Å, well beyond the 2.85–3.40 Å separations observed for these interactions.

Another possibility is suggested by the electrostatic potential on the molecular surface of nitromethane (the latter being defined by the 0.001 au contour of the electron density⁵³). The surface is divided into a large positive region and a slightly smaller negative one, the latter being associated with the oxygens of the nitro group. The key point is that the potential in each of these two regions varies to a lesser degree than in many other polar molecules; the maxima and minima are less pronounced. Quantitative indicators of this are the variances of the positive and negative potentials, σ_+^2 and σ_-^2 , which are statistical measures of their ranges and variabilities.⁵⁴ Their computed magnitudes for the nitromethane molecule, as calculated at the density functional B3P86/6-31+G** level, are $\sigma_+^2 = 58.2$ kcal²/mol², $\sigma_-^2 = 62.2$ kcal²/mol². In contrast, the analogous quantities for water are 192.7 and 119.3 kcal²/mol², and for methanol, they are 94.5 and 134.2 kcal²/mol². It follows from these data that while an electrostatic attraction between nitromethane molecules is certainly to be anticipated, it should be less site-specific and less directional, as well as weaker, than those between water or methanol molecules. Thus, while it may be argued that there is a sort of hydrogen bonding in liquid nitromethane at 1 atm, it is certainly more diffuse, less oriented, and less persistent than in water or methanol. This conclusion is fully consistent with the analyses of orientational dynamics discussed above.

Summary

The results of this initial investigation of a potential function and simulation procedure for liquid nitromethane are overall satisfactory, although there is room for improvement (e.g., in the dielectric constant). Some avenues to explore include the introduction of anharmonicity in the intramolecular potential terms, the introduction of polarization in the intermolecular terms,^{55,56} modifications of the parametrization schemes, longer simulation time in determining the dielectric constant, larger system size, etc. The calculated reorientation times and the molecular surface electrostatic potential indicate that the intermolecular interactions in liquid nitromethane are not highly directional and site-specific, as would be expected if strong hydrogen bonding were involved.

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