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Relaxation and transport properties of liquid n-triacontane

N D Kondratyuk, A V Lankin, G E Norman and V V Stegailov

Joint Institute for High Temperatures of the Russian Academy of Sciences, Izhorskaya 13 Bldg 2, Moscow 125412, Russia

E-mail: kondratyuk@phystech.edu

Abstract. Molecular modelling is used to calculate transport properties and to study relaxation of liquid n-triacontane ($C_{30}H_{62}$). The problem is important in connection with the behavior of liquid isolators in a pre-breakdown state. Two all-atom models and a united-atom model are used. Shear viscosity is calculated using the Green–Kubo formula. The force fields are compared with each other using the following criteria: the required time for one molecular dynamics step, the compliance of the main physical and transport properties with experimental values. The problem of the system equilibration is considered. The united-atom potential is used to model the n-triacontane liquid with an initial directional orientation. The time of relaxation to the disordered state, when all molecules orientations are randomized, are obtained. The influence of the molecules orientations on the shear viscosity value and the shear viscosity relaxation are treated.

1. Introduction

Modern industry is strongly interested in studying of liquid hydrocarbon properties, since they are part of the lubricants, insulation and fuel liquids. Optimization of these properties and their dependences on external parameters in such liquids are long-standing matters of research.

Molecular dynamics (MD) method is a fundamental research tool that opens the possibility to study various systems (e.g. [1,2]) using high performance computing [3]. There are molecular dynamics studies of the liquid alkanes (up to $C_{16}H_{34}$) [4–7]. Transferable Potential for Phase Equilibria (TraPPE) [4] for hydrocarbons is mostly used in many of them. It is applied for predicting structural and thermodynamic behavior of n-hexane, n-decane and n-heptadecane in [5]. The shear viscosity calculations for n-decane and n-hexadecane are performed in [6]. The physical properties of the liquid n-hexane are obtained in [7].

MD is used to understand the relation between the chemical structure and lubricant performance of liquid C_{30} isomers [8]. The shear viscosity is calculated for n-triacontane and two branched alkanes using the Rouse model. The correspondence with experiment within accuracy of 15% is found for the branched alkane squalane ($C_{30}H_{50}$). However, the results for the n-triacontane show about 50% error with respect to the experimental data. The calculation of transport properties for the n-triacontane is actual, because this alkane is a part of transformer oils. It is the subject of this work.

All-atom approach becomes more applicable due to the development of computing power in recent years. DREIDING (named after André Dreiding) [9] force field is used for prediction of thermal and mechanical properties for epoxy polymer [10, 11]. Optimized Potential for Liquid



Simulations (OPLS) [12] is used in [13] to simulate self-diffusion and structural properties of n-alkanes up to n-decane. All-atom models provide more accurate simulations in most cases (although they are certainly less accurate than quantum chemical methods, e.g. see [14]).

Liquid n-triacontane $C_{30}H_{62}$ is taken for MD simulation study in this paper. The molecule is schematically shown in figure 1. It is one of the important representatives of n-alkanes which are the main components of transformer oils. The molecule $C_{30}H_{62}$ is long enough for the orientations effects and rheological properties to be studied. The basic relations, the model and the simulation technique are given in sections 2, 3 and 4 correspondingly. The shear viscosity is obtained through the Green–Kubo formalism in 5.1. The values of the physical properties in the TraPPE, DREIDING and OPLS force fields are compared in 5.2. The case, when all molecules are oriented in one direction, is also treated. The influence of this ordering on the viscous properties and conformational relaxation are studied in 5.3. This situation has not been discussed on the molecular level yet. It could take place, when molecules are in a strong electric field. The behavior of the isolators in a pre-breakdown state is treated at the macro level in [15,16].

2. Basic relations

The viscosity $\eta_{\alpha\beta}$ is calculated using the Green–Kubo formula [17]:

$$\eta_{\alpha\beta} = \frac{V}{k_B T} \int_0^{t'} C_\sigma(t) dt, \quad (1)$$

$$C_\sigma(t) = \langle \sigma_{\alpha\beta}(0) \sigma_{\alpha\beta}(t) \rangle, \quad (2)$$

where $C_\sigma(t)$ is an autocorrelation function, $\sigma_{\alpha\beta}$ are off-diagonal components of the stress tensor, V and T are system volume and temperature, k_B is Boltzmann's constant. $\langle \dots \rangle$ in equation (2) is an average over the canonical ensemble. t' in (1) is a value of time when the autocorrelation function asymptotically decays to zero. The shear viscosity η is found as an average of the η_{xy} , η_{xz} and η_{yz} .

The stress tensor $\sigma_{\alpha\beta}$ is calculated from the next equation:

$$\sigma_{\alpha\beta} V = \sum_{i=1}^N m_i v_{i\alpha} v_{i\beta} + \sum_{i=1}^N r_{i\alpha} f_{i\beta}, \quad (3)$$

where N is a number of atoms, $r_{i\alpha}$ and $v_{i\alpha}$ are α -components of coordinate and velocity of the i -th atom, and $f_{i\alpha}$ is α -component of the force that acts on the i -th atom.

3. Model

3.1. Force fields

The force fields, which are used in this work, include intramolecular and intermolecular parts. The intramolecular part describes interactions between atoms of the same molecule. It contains Lennard-Jones, Coulomb and bonded, as well as angle and torsion interactions (figure 2):

$$E = E_{LJ} + E_{Coul} + E_{bond} + E_{angle} + E_{torsion}. \quad (4)$$

The bonded and angle interactions have harmonic forms:

$$E_{bond} = K_b (R - R_0)^2, \quad (5)$$

$$E_{angle} = K_a (\theta - \theta_0)^2, \quad (6)$$

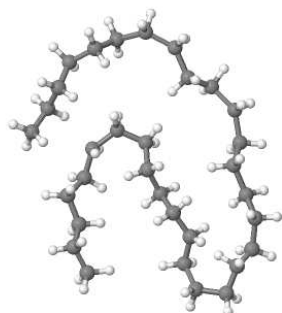


Figure 1. n-triacontane molecule $C_{30}H_{62}$.

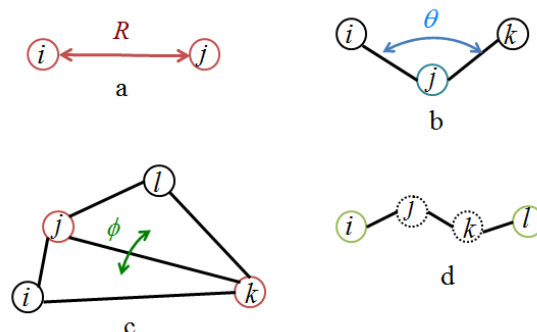


Figure 2. Bonded (a), angle (b), torsion (c) and Lennard-Jones (d) types of interaction.

where K_a and K_b are energy constants, R and θ are a bond-length and an angle, R_0 and θ_0 are an equilibrium bond-length and an angle. The Lennard-Jones term has a 6-12 form:

$$E_{LJ} = 4\epsilon \left[(\sigma/r)^{12} - (\sigma/r)^6 \right], \quad (7)$$

where ϵ is an energy constant, σ is a zero-crossing parameter, r is a distance between two atoms. Non-bonded interactions in the molecule (between atoms, that are not included in the bonded and angle interactions) are also represented via the Lennard-Jones form. The intermolecular part describes forces between atoms that belong to different molecules. It consists of Lennard-Jones and Coulumb parts.

DREIDING includes all atoms of the system. The torsion interaction has a form:

$$E_{torsion} = K_{JK}(1 - \cos[n_{JK}(\phi - \phi_{JK}^0)]), \quad (8)$$

where K_{JK} is a barrier to rotation, n_{JK} is a periodicity, ϕ_{JK}^0 is an equilibrium angle. The parameters depend on the hybridization of J and K atoms. J and K are sp-3 hybridized atoms in the case of n-triacontane.

OPLS is also an all-atom model. The difference is that it includes Coulumb interaction Cq_iq_j/r , where q_i and q_j are charges of the i -th and j -th atoms, r is a distance between them. The changes in the distribution of the electron density (due to the polar covalent bonds inside molecules) create non-zero atomic charges. Carbon atoms have $-0.18 e$ in the methyl group and $-0.12 e$ in the methylene group. Hydrogen atoms have $+0.06 e$ in all groups. OPLS uses a more complex form of torsion interaction:

$$2E_{torsion} = K_1(1 + \cos(\phi)) + K_2(1 - \cos(2\phi)) + K_3(1 + \cos(3\phi)) + K_4(1 - \cos(4\phi)), \quad (9)$$

where K_1 , K_2 , K_3 , K_4 are energy constants, ϕ is a dihedral angle.

TraPPE is a united-atom potential. Methyl (CH_3) and methylene (CH_2) groups are represented as two point particles in the case of this force field. Positively charged hydrogen atoms are merged with an electronegative carbon atom in each group. All groups have zero charge and Coulomb interactions are excluded due to this fact. TraPPE uses the same formulas as (5)–(7), (9), but the values of parameters are different. The number of constants is less and therefore the parametrization becomes simpler.

TraPPE model requires three times less particles than all-atom models to simulate the alkane liquid. Therefore it needs the least computational time per an MD step.

3.2. Conformation analysis

The process of relaxation is nontrivial to be controlled in a system of long molecules [18]. It requires the use of additional criteria. The relative shape anisotropy κ^2 is applied in this paper. This parameter is used for a conformational analysis of large proteins and biomolecules [19]. It shows how atoms are located relative to the center of mass of the molecule:

$$\kappa^2 = \frac{3}{2} \frac{\lambda_1^4 + \lambda_2^4 + \lambda_3^4}{(\lambda_1^2 + \lambda_2^2 + \lambda_3^2)^2} - \frac{1}{2}, \quad (10)$$

where λ_i ($i = 1, 2, 3$) are eigenvalues of the inertia tensor:

$$I_{\alpha\beta} = \frac{1}{m} \sum_{i=1}^{N_a} m_i (r_{i\alpha} - r_{cm\alpha})(r_{i\beta} - r_{cm\beta}), \quad (11)$$

where $r_{i\alpha}$ is an α -component of the radius vector of the i -th atom, m and $r_{cm\alpha}$ are a molecule mass and an α -component of center of mass coordinate, N_a is a number of atoms in a molecule. The κ^2 value lies in the interval $[0, 1]$.

If $\kappa^2 = 1$ the molecule is planar. If $\kappa^2 = 0$ atoms in the molecule are spherically symmetrical about the center of mass. If the system is equilibrated the average value of κ^2 becomes a constant.

4. Simulation technique

4.1. Equations of motion

The dynamics of N particles is described by the second Newton's law:

$$m_i d^2 \vec{r}_i / dt^2 = \vec{F}_i(r_1, \dots, r_N), \quad (12)$$

where m_i , \vec{r}_i , \vec{F}_i are a mass, a radius vector and a force of the i -th atom. Or it can be treated as:

$$m_i d\vec{v}_i / dt = \vec{F}_i(\vec{r}_1, \dots, \vec{r}_N), \vec{v}_i = d\vec{r}_i / dt,$$

where \vec{v}_i is a velocity of the i -th atom. The force is described as:

$$\vec{F}_i = -\partial E(r_1, \dots, r_N) / \partial \vec{r}_i, \quad (13)$$

where $E(\vec{r}_1, \dots, \vec{r}_N)$ is an interaction potential (4), which determines the dynamics of the system.

The Velocity-Verlet algorithm is used to solve equations (12):

$$\begin{aligned} \vec{v}_i(t + \Delta t/2) &= \vec{v}_i(t) + \vec{F}_i(t) \Delta t / 2m_i, \\ \vec{r}_i(t + \Delta t) &= \vec{r}_i(t) + \vec{v}_i(t + \Delta t/2) \Delta t, \\ \vec{v}_i(t + \Delta t) &= \vec{v}_i(t + \Delta t/2) + \vec{F}_i(t + \Delta t) \Delta t / 2m_i. \end{aligned}$$

Molecular dynamics simulations are carried out using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) package [20].

4.2. Statistics

Calculation results should be obtained by averaging over an MD trajectory. Therefore, the trajectory should be divided into statistically independent parts. They should be about the value of the memory time t_m . t_m is a time during which the system forgets its initial conditions. It can be obtained from the asymptotic forms of the following equations [18]:

$$\langle \Delta r^2 \rangle = \frac{1}{N} \sum_{i=1}^N (r_i(t) - r'_i(t))^2, \langle \Delta v^2 \rangle = \frac{1}{N} \sum_{i=1}^N (v_i(t) - v'_i(t))^2, \quad (14)$$

where $(r;v)(t)$ and $(r';v')(t)$ are the trajectories with the same initial conditions, integrated using Δt and $\Delta t'$ timesteps. The asymptotes of these divergences are $\langle \Delta r^2 \rangle = A \exp(Kt)$, $\langle \Delta v^2 \rangle = B \exp(Kt)$, if $t < t_m$. $\langle \Delta r^2 \rangle = 2D(t - t_m)$, $\langle \Delta v^2 \rangle = 2v_{th}^2$, if $t > t_m$, where D is a diffusion coefficient, $v_{th} = \sqrt{3k_B T/m}$. The behavior of these divergences is studied for ionic liquids in [21]. The results of that work show that anomalous diffusion ($\langle \Delta r^2 \rangle \sim t^\alpha$, $\alpha < 1$) takes place after the ballistic regime in the case of large molecules. Therefore the diffusion coefficient D becomes a function of time and should be obtained from the long trajectory.

The divergences (14) are calculated for molecules centers of mass using two trajectories integrated with $\Delta t = 0.5$ fs and $\Delta t = 1$ fs (figure 3). $\langle \Delta r^2 \rangle$ is normalized to the squared average distance between molecules $n^{-2/3}$. The memory time is defined as a moment, when linear and constant asymptotes of $\langle \Delta v^2 \rangle / 2v_{th}^2$ cross with each other. The obtained value is 2 ps.

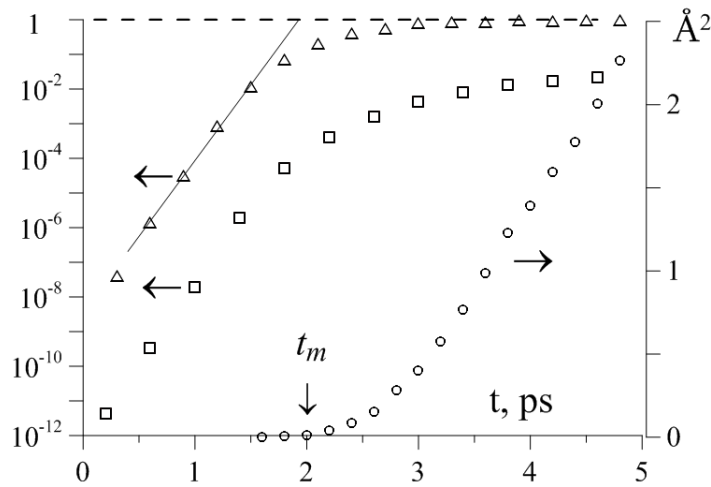


Figure 3. Molecules centers of mass divergences (TraPPE). Triangles are $\langle \Delta v^2 \rangle / 2v_{th}^2$. Squares are $\langle \Delta r^2 \rangle n^{2/3}$. Circles are $\langle \Delta r^2 \rangle$. Trajectories for (14) are integrated with 0.5 and 1 fs timesteps.

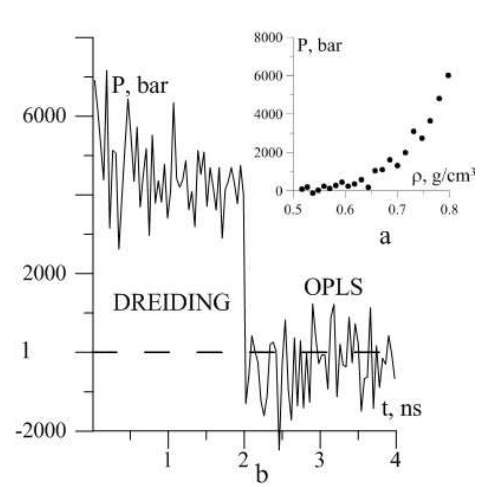


Figure 4. a) Density dependence on pressure in the DREIDING model. b) The switching between DREIDING and OPLS force fields.

The values of the autocorrelation function inside each interval are calculated in LAMMPS as: $\langle \sigma_{\alpha\beta}(0)\sigma_{\alpha\beta}(\delta) \rangle = [\sigma_{\alpha\beta}(0)\sigma_{\alpha\beta}(\delta) + \sigma_{\alpha\beta}(\delta)\sigma_{\alpha\beta}(2\delta) + \dots + \sigma_{\alpha\beta}(t_{int} - \delta)\sigma_{\alpha\beta}(t_{int})] / M$, where δ is a moment when the correlation is calculated, t_{int} is a length of the one trajectory interval, M is a number of such points inside this interval ($M \sim 10^4$). The next step is an averaging over these trajectory parts.

The authors [6] use another approach for calculating $\langle \sigma_{\alpha\beta}(0)\sigma_{\alpha\beta}(t) \rangle$ for n-hexadecane. The result is averaged over independent trajectories, but the length of each trajectory is shorter. They are generated using a variety of parameters of the relaxation to the initial configuration. It is not necessary if there is enough computational resources to run a long MD trajectory.

4.3. Relaxation to the initial configuration

The starting configuration which we use in the TraPPE model is a gas of 125 replicated n-triacontane molecules. The distance between them is longer than the force field cutoff radius (14 Å). At the first step temperature is set to 600 K. This temperature is needed to perform molecules disordering quickly. Molecules get random orientation in the NVE ensemble for 0.1 ns. The integration timestep is 1 fs. The second step is compression to the experimental density 0.775 g/cm^3 [22] for 0.1 ns. The third step is relaxation in the NPT thermostat for 2 ns. The average density at the last 0.5 ns is chosen as the equilibrium. It corresponds to the experimental value which is available at 353 K. This temperature is achieved in the NPT ensemble. The last step is relaxation in the NVT ensemble at the calculated density for 2 ns.

The generation of the initial configuration is slightly different in the DREIDING model. The first two steps are the same. The pressure is overestimated at the experimental density. Therefore, relaxation in the NPT thermostat leads to the rarefied liquid that does not correspond to the real behavior under these conditions. The third step is skipped because of this fact. The last step is relaxation only in the NVT ensemble for 3 ns at the experimental density, the parameters are the same as in the TraPPE model.

5. Results

MD calculations are carried out for trajectories starting from equilibrated initial configurations, that are obtained in the previous section 4.3.

5.1. Force field comparison

DREIDING does not reproduce the equation of state correctly. The pressure is very high at the physical density. A small change in density could not correct this situation, because the pressure begins increasing at 0.6 g/cm^3 (figure 4a). It is compared with OPLS to understand how the parametrization of the total energy (4) influences the pressure.

The MD trajectory is integrated in the NVE ensemble. DREIDING is changed to OPLS at 2 ps (figure 4b) and relaxation of the main properties is studied. The pressure becomes normal in the OPLS model. Fluctuations result from the smallness of the system.

The total energy also becomes lower. The Coulomb interaction and the more complicated torsion form give the difference in the physical properties.

The average relative shape anisotropy $\langle \kappa^2 \rangle$ is not changed. It means, that the configurations of the system in both models are the same.

OPLS increases significantly the computational time required for one MD step.

TraPPE also provides the accurate description of the physical properties (both pressure and structure). And it takes less time for the MD step due to united-atom approach. Therefore TraPPE is efficient for the system equilibration stage.

5.2. Shear viscosity

The autocorrelation function $C_\sigma(t)$ smoothly decays to zero in atomic systems, the time, when it becomes asymptotically zero, is about 1 ps [23].

It appears to oscillate in this study (figure 5). The existence of bonds inside the molecules is the reason of the unusual behavior. They contribute to the calculation of the stress tensor [24]. The oscillation frequency is lower in the united-atom model, because C–H bonds are excluded.

The shear viscosity in the TraPPE model is calculated in the 4 ns run. The trajectory is divided into parts of $t_{int} = 15 \text{ ps}$. The autocorrelation function is averaged and integrated from 0 to $t' = 15 \text{ ps}$ on each part. These running integrals (1) are shown in figure 6. The function collects statistics at the first 1500 ps and then the integral value becomes a constant.

The obtained value is $0.96 \text{ mPa}\cdot\text{s}$, which is lower than the experimental value $4.87 \text{ mPa}\cdot\text{s}$ (table 1). The number of atoms in the united-atom model is much less than in the all-atom

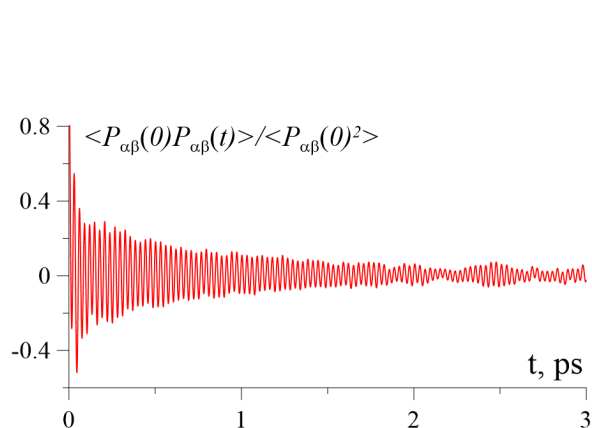


Figure 5. The behavior of the normalized stress-stress correlation function in the TraPPE model.

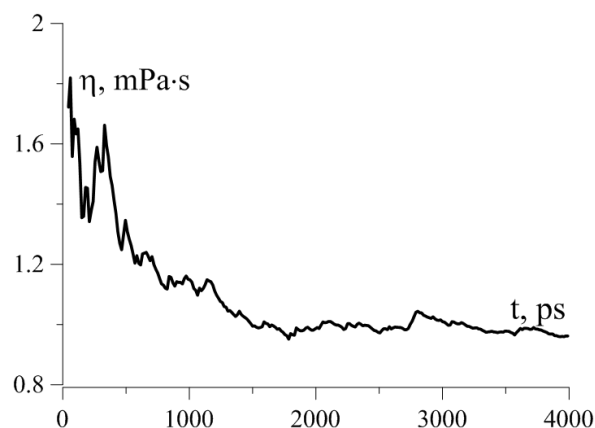


Figure 6. The running shear viscosity calculation with $t_{int} = 15$ ps, $t' = 15$ ps in the TraPPE force field.

model at the same density. Molecules have more free space to move and therefore there is less friction. This is the reason of underestimated viscosity coefficient in the TraPPE force field.

The calculations with the same parameters are provided in the DREIDING force field. But the time of the full trajectory is 3 ns, because the MD step requires more computational time. The obtained value is 4.33 mPa·s. The all-atom model gives a better agreement with experimental data.

Table 1. Physical properties for the different models at 353 K and 0.775 g/cm^3 .

Model	TraPPE	DREIDING	OPLS	Exp
Pressure, bar	1	4200	1	1
Shear viscosity, mPa·s	0.96	4.33	—	4.87 [22]

5.3. Ordered system

Polarized molecules should become oriented in the prebreakdown state, because they have dipole moments. This fact can influence the prebreakdown state and change breakdown conditions in comparison with unpolarized liquid.

Critical situation (most of the molecules are oriented in one direction) is simulated to understand the maximum possible influence on the shear viscosity. The modelling is carried out in the TraPPE force field.

The generation of the initial configuration is performed by changing parameters of the intramolecular part. The Lennard-Jones forces between methylene particles, which are separated by two other particles, are called the 1-4 interaction. If the 1-4 interaction is zeroed, it gives additional rigidity to the molecule. This change is applied during the whole process of creating of the initial configuration.

The first step is a gas of the oriented molecules in x direction. The second step is a compression to the experimental density for 0.5 ns. Clusters with a preferred orientation are formed during the compression. All the clusters are merged at the end of the process.

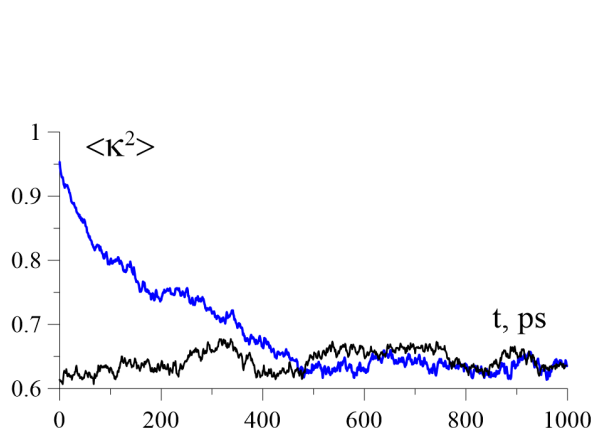


Figure 7. The dynamics of $\langle \kappa^2 \rangle$. Blue line is the relaxation. Black line corresponds to the disordered liquid.

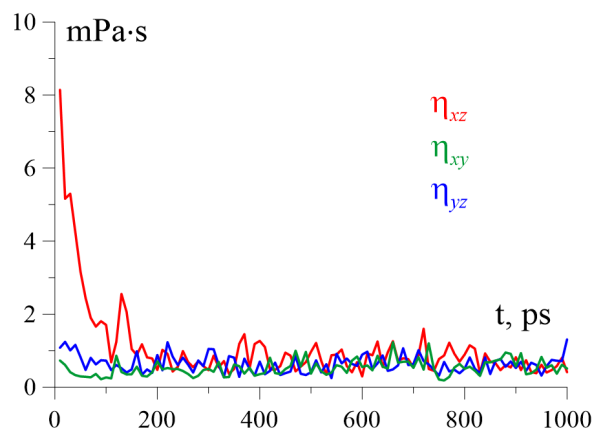


Figure 8. The running shear viscosity calculation during relaxation. $t' = 3$ ps, $t_{int} = 10$ ps. The average value for the each direction is taken over 10 independent trajectories.

The temperature of the initial configuration is set to 400 K. The value of η_{xz} increases by order of magnitude because of the molecules polarization (figure 8). $\langle \kappa^2 \rangle$ value is about 1, which means that all the molecules are oriented and extended.

The time of relaxation is too small to collect correct statistics for the autocorrelation function. Therefore, the average in the equation (2) should be taken over the independent trajectories, which are generated with different initial conditions. The random number generator takes a seed to create a velocity distribution in the beginning of the relaxation. 10 different seeds are used to generate statistically independent velocity distributions.

The average relative shape anisotropy $\langle \kappa^2 \rangle$ is calculated during the relaxation (figure 7). It decreases to the value 0.63 that corresponds to the disordered liquid. The time of conformational relaxation to the disordered system is 500 ps.

The viscosity isotropy is violated in this case. The viscosity value in xz plane becomes equal to xy, yz during the process of relaxation. The time of this process is 200 ps.

6. Conclusions

MD is used to calculate the shear viscosity value of the liquid n-triacontane in the different force fields. The influence of the molecule orientations on the viscosity coefficient in different directions is treated.

1) DREIDING, OPLS and TraPPE are compared by the pressure dependence on density that they reproduce. The last two models give realistic values of pressure at the physical density, in the DREIDING force field pressure is overestimated.

2) TraPPE can be used for producing initial configurations quickly, because it takes the least computational time for one MD step. It can be changed to all-atom models for obtaining more accurate values of transport properties.

3) The shear viscosity is calculated in the TraPPE and DREIDING force fields. The value in the first force field is low (−80%) because of the united-atom approach. This model is good for simulating thermodynamics and initial configurations of the liquid alkanes. In the second model the value is more accurate (−11%). It is better to use all-atom models to reproduce and predict transport properties of alkanes.

4) The situation when all molecules are oriented in one direction is studied. The effect of the viscosity anisotropy appears in this case. Shear viscosity coefficient in the preferred

direction increases by an order of magnitude and then relaxes to the normal value. The time of viscosity relaxation is 200 ps. The average relative shape anisotropy is useful for detecting of the conformational relaxation. The time, when the system gets its average conformation, is 500 ps.

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