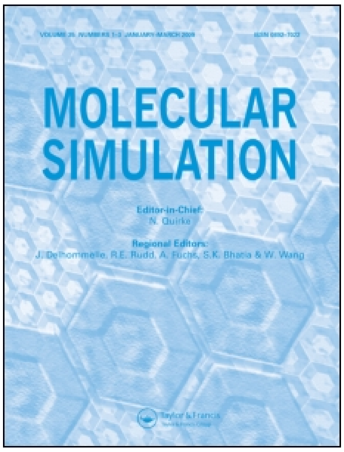

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# Thermal Conductivity of Continuous, Semi-continuous and Discrete Mixtures using Nonequilibrium Molecular Dynamics

P. Ravi[a]; S. Murad[a]
[a] Chemical Engineering Department, University of Illinois at Chicago, Chicago, IL, U.S.A.

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# THERMAL CONDUCTIVITY OF CONTINUOUS, SEMI-CONTINUOUS AND DISCRETE MIXTURES USING NONEQUILIBRIUM MOLECULAR DYNAMICS

P. RAVI and S. MURAD

*Chemical Engineering Department, University of Illinois at Chicago, Chicago, IL 60680-4348, U.S.A.*

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The thermal conductivity of continuous (or polydisperse), semi-continuous and discrete dense fluid mixtures has been investigated using the nonequilibrium molecular dynamics method of Evans [1]. Linear, gaussian and unsymmetric distributions have been studied. The results have been used to examine the *n*-component characterizations for such mixtures and the effect of the various distributions on these characterizations. We show that the 2-component characterization of these mixtures is significantly better than the 1-component characterization and that going to higher order characterizations does not improve results any further. In addition, equimolar characterizations are found to give better results than equiparameter (e.g. $\sigma$, $\varepsilon$) characterizations, in the case of unsymmetric distributions.

KEY WORDS: Thermal conductivity, polydisperse, fluid mixtures, nonequilibrium molecular dynamics.

## INTRODUCTION

The accurate estimation of transport properties of many component mixtures (also referred to as polydisperse or continuous) is an important scientific and engineering problem. Often fluids encountered in various chemical processes, e.g., petroleum reservoir fluids, coal liquids, vegetable oils, etc. are continuous [2]. To study such mixtures, one generally models them as *n*-component hypothetical mixtures (the so called *n*-fluid characterizations) using suitable mixing rules. After such a characterization, engineering correlations can be used to predict their transport properties [3].

In this paper we investigate the thermal conductivity of many component mixtures using the nonequilibrium molecular dynamics (NEMD) method of Evans [1]. Several types of mixtures have been studied, including continuous, semi-continuous and discrete mixtures. The results have been used to assess the accuracy of the *n*-component characterizations of these many component mixtures.

## THEORY AND METHOD

In our work we have used the non-canonical linear response theory method [1] to investigate thermal conductivity. In this method an external field is used to increase the energy of all particles in the system with more than the average energy, and decrease it for those with less than the average energy. This leads to a heat flux in the system, whose magnitude is used to estimate the thermal conductivity. The equations of motion given below show how such an external field can be incorporated into the equations for an NEMD study, for a potential model $\phi_{ij}$ [1].

$$\dot{\mathbf{r}}_i = \mathbf{p}_i/m \tag{1}$$

$$\dot{\mathbf{p}}_i = \sum_j \mathbf{f}_{ij} + (E_i - \bar{E})\mathbf{F}(t) + \frac{1}{2}\sum_j \mathbf{f}_{ij}\,[\mathbf{r}_{ij}\cdot\mathbf{F}(t)] - \frac{1}{2N}\sum_{jk} \mathbf{f}_{jk}\,[\mathbf{r}_{jk}\cdot\mathbf{F}(t)] - \alpha\mathbf{p}_i \tag{2}$$

where $\mathbf{f}_{ij} = -\partial\phi_{ij}/\partial\mathbf{r}_i$, $\mathbf{r}_i$ is the position, $m$, the mass, $\mathbf{p}_i$, the momentum, and $E_i$, the energy of particle $i$ and $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$. $\bar{E}$ is the average energy per particle and $N$ is the number of particles in the system. $\mathbf{F}(t)$ is the external field which is kept constant and $\alpha$ (the thermostatting multiplier) ensures isothermal conditions. The thermal conductivity $\lambda$, can then be estimated from the equation,

$$\lambda = \frac{1}{T}\frac{\langle \mathbf{J}_Q(t\to\infty)\rangle}{\mathbf{F}(t)} \tag{3}$$

where $T$ is the temperature and the heat flux vector is

$$\mathbf{J}_Q = \frac{1}{V}\left[\sum_i E_i\frac{\mathbf{p}_i}{m} + \frac{1}{2}\sum_{ij}\mathbf{r}_{ij}\left(\frac{\mathbf{p}_i}{m}\cdot\mathbf{f}_{ij}\right)\right] \tag{4}$$

Equations (1–4) can be extended to mixtures in a straightforward manner. However, one must also account for the heat flux due to diffusion as shown below [4].

$$\mathbf{J}_Q = \frac{1}{V}\left[\sum_i E_i\frac{\boldsymbol{\pi}_i}{m_i} + \frac{1}{2}\sum_{ij}\mathbf{r}_{ij}\left(\frac{\boldsymbol{\pi}_i}{m_i}\cdot\mathbf{f}_{ij}\right)\right] \tag{5}$$

In Equation (5) $\mathbf{p}$ has been replaced with $\boldsymbol{\pi}$ to account for diffusive heat flux and is defined as,

$$\boldsymbol{\pi}_i = \mathbf{p}_i - \frac{1}{N_v}\sum^{v}\mathbf{p}_i \tag{6}$$

$V$ is the volume of the simulation cube, and $N_v$ is the number of particles of species $v$ and the summation in Equation (6) is only over species $v$.

The intermolecular potential, $\phi$, we have used in this work is the Lennard-Jones potential. We would like to note that transport properties such as thermal conductivity are known not to be very sensitive to the form of the intermolecular potential used and the results obtained here should be realistic for a wide range of compounds [5]. This has been confirmed recently for the thermal conductivity of HCl, where we obtained essentially similar results, using four different potential models [6]. For mixtures we have used the usual Lorentz–Berthelot rules for cross interactions [5].

Several mixing rules have been developed in the past few decades. These mixing rules can be used either for molecular parameters, such $\varepsilon$, $\sigma$, $M$ ($\varepsilon$ and $\sigma$ are the Lennard-Jones energy and size parameters and $M$ is the molecular weight) or for the property of interest, such as thermal conductivity, directly. In the first case, the molecular parameters of the constituents of the mixture are used to obtain these parameters for the hypothetical mixture ($\varepsilon_x$, $\sigma_x$, $M_x$); the property of interest (thermal conductivity) of such a mixture (with parameters $\varepsilon_x$, $\sigma_x$, $M_x$) can then be calculated using any appropriate method, such as NEMD. In the second case, the thermal conductivity of the constituents can be used to obtain the thermal conductivity of the mixture directly. In our work we have used these mixing rules for the molecular parameters needed to characterize the mixtures only. In a previous study [7] we have shown that for the thermal conductivity of dense fluid mixtures, the simple Kay's mixing rules for molecular parameters do generally as well as the more sophisticated Enskog rules. Therefore all the mixtures studied here have been characterized using Kay's rules. Kay's mixing rules are given by,

$$\begin{aligned} \sigma_x &= \sum_\alpha x_\alpha \sigma_\alpha \\ \varepsilon_x &= \sum_\alpha x_\alpha \varepsilon_\alpha \\ M_x &= \sum_\alpha x_\alpha M_\alpha \end{aligned} \tag{7}$$

The NEMD algorithm described earlier was used to calculate the thermal conductivity of many component mixtures. The details of the simulation technique used are described in the Appendix. Values of $\mathbf{F}(t)$ (Equation (2)), were chosen so that $\lambda$ (Equation (3)) was independent of $\mathbf{F}(t)$. To simplify calculations, we applied $\mathbf{F}(t)$, along the $z$-coordinate. These mixtures were then characterized via the Kay's rules as a 1-, 2-, 4-, 8- and 16-component hypothetical mixtures (see discussion following Figure 1). The thermal conductivity of these hypothetical $n$-component mixtures (whose molecular parameters were obtained from Kay's rules) was calculated using the NEMD method. The results obtained enabled us to examine the accuracy of the various $n$-component characterizations.

## RESULTS

We examined mixtures having various distributions with molecular sizes between $\sigma_1$ and $\sigma_2$, molecular weights between $M_1$ and $M_2$, and energy parameters between $\varepsilon_1$ and $\varepsilon_2$. The smallest and largest values of these molecular parameters roughly correspond to those in the $C_1/C_{16}$ hydrocarbon range. Figure 1 shows a linear and two gaussian distributions with molecular sizes between $\sigma_1$ and $\sigma_2$. The cumulative mole fraction, $x$, for the linear distribution is given by,

$$x(\sigma) = (\sigma - \sigma_1)/(\sigma_2 - \sigma_1) \tag{8}$$

while for the gaussian distributions it is given by,

$$x(\sigma) = \int_a^y e^{-t^2/2}\,dt \Big/ \int_a^b e^{-t^2/2}\,dt \tag{9}$$

where $a = (\sigma_1 - m)/s$, $b = (\sigma_2 - m)/s$, and $y = (\sigma - m)/s$. $m$ is the mean and $s$ is the standard deviation of the distribution. Gaussian I has a mean of $(\sigma_2 + \sigma_1)/2$ and a standard deviation of $(\sigma_2 - \sigma_1)/4$ and Gaussian II has the same mean but a standard deviation of $(\sigma_2 - \sigma_1)/8$, i.e., half that of Gaussian I.

The mixtures were characterized as 1-, 2-, 4-, 8- and 16-component equimolar mixtures. Figure 1 shows a 4-component characterization of the molecular parameter $\sigma$, for the linear distribution case using Kay's mixing rule. In this case one can characterize all components with $x_i \leqslant 0.25$ as one component and so on. We have

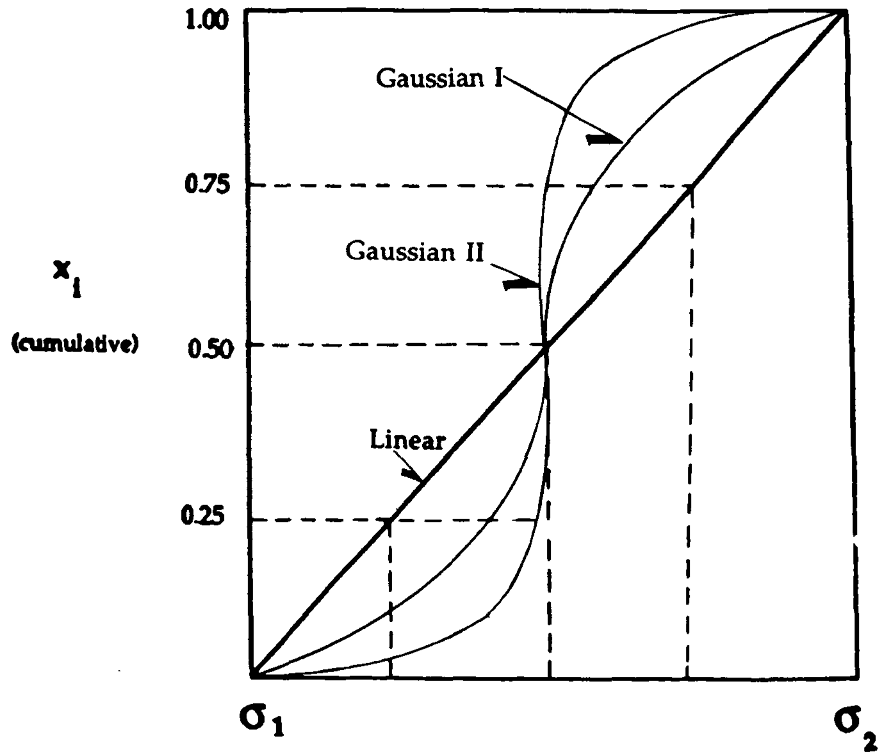

**Figure 1** Characterization of a continuous mixture with a finite number of components using Kay's mixing rule. An example of a 4-component equimolar characterization is shown for the linear distribution case. Here all components with $x_i \leq 0.25$ are characterized as one component and so on. Also shown are two gaussian distributions.

carried out studies in which mainly the molecular size ratio varies, since this is expected to be the most important parameter for thermal conductivity. The energy parameter ratio and the molecular mass ratio were also varied for some distributions. Both high and low densities were studied. We estimate the accuracy of each individual simulation made to obtain the thermal conductivity of these mixtures, to be within 4 percent. *All results reported in this paper were obtained using NEMD.* The $\lambda$ values are reported in reduced units as, $\lambda^* = \lambda\sigma_1^2 m_1^{1/2}/k\,\varepsilon_1^{1/2}$.

Figure 2 shows typical results obtained when a continuous mixture was modelled as 1-, 2-, 4-, 8- and 16-component equimolar hypothetical mixtures. Note that the Kay's mixing rules are used only to obtain the molecular parameters for the hypothetical mixtures (1-, 2-, 4-component, etc.), and NEMD to obtain the thermal conductivity of these $n$-component mixtures directly. $\lambda_0$, the thermal conductivity of the continuous mixture, has been obtained by extrapolating results for mixtures modelled by upto 16 components. In view of Figure 2, this should be entirely satisfactory. It can be clearly seen that the biggest change (improvement) comes in going from a 1-component characterization, to a 2-component characterization. Going to higher order

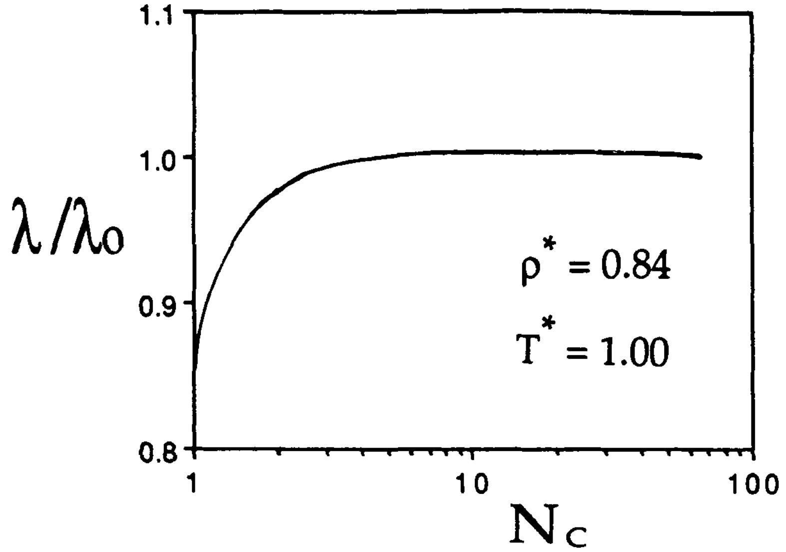

**Figure 2** Effect of number of components ($N_c$) used in characterizing continuous mixtures on thermal conductivity ($\lambda$) for linear distributions, showing the improvement in going from one to two components. $\lambda_0$ is the thermal conductivity of the continuous mixture.

characterizations does not improve results. This was found to be true for all the distributions examined in this paper.

Table 1 shows results for continuous mixtures with linear distributions. Results are shown for cases where the size, energy and molecular mass ratios are varied individually and all three parameters simultaneously. $\lambda_0$ is the thermal conductivity of the continuous mixture obtained as shown in Figure 2. $\lambda_2$ is the thermal conductivity of the 2-component characterization of this continuous mixture. Similarly $\lambda_1$, is the 1-

**Table 1** Thermal conductivity of continuous mixtures with linear distributions.

| $\varepsilon_2/\varepsilon_l$ | $\sigma_2/\sigma_l$ | $M_2/M_l$ | $\rho^{*a}$ | $T^{*a}$ | $\lambda_0$ | $\lambda_2$ | $\lambda_l$ |
|---|---|---|---|---|---|---|---|
| 1.0 | 0.5 | 1.0 | 0.42 | 1.0 | 3.90 | 3.90 | 4.33 |
| 1.0 | 0.7 | 1.0 | 0.40 | 1.0 | 2.72 | 2.72 | 2.94 |
| 1.0 | 1.5 | 1.0 | 0.78 | 1.0 | 3.99 | 3.99 | 3.83 |
| 1.0 | 2.0 | 1.0 | 0.68 | 1.0 | 2.14 | 2.08 | 1.90 |
| 1.0 | 2.0 | 1.0 | 0.84 | 1.0 | 3.91 | 3.83 | 3.36 |
| 1.0 | 3.0 | 1.0 | 0.80 | 1.0 | 2.48 | 2.36 | 1.66 |
| 1.0 | 1.0 | 5.0 | 0.84 | 1.0 | 4.00 | 4.12 | 4.08 |
| 1.0 | 1.0 | 8.0 | 0.84 | 1.0 | 3.04 | 3.10 | 3.47 |
| 1.0 | 1.0 | 10.0 | 0.84 | 1.0 | 2.73 | 2.73 | 3.28 |
| 1.0 | 1.0 | 10.0 | 0.42 | 1.0 | 0.87 | 0.90 | 0.95 |
| 4.0 | 1.0 | 1.0 | 0.84 | 1.0 | 10.96 | 11.40 | 11.62 |
| 4.0 | 2.0 | 10.0 | 0.84 | 1.0 | 2.35 | 2.35 | 2.26 |
| 4.0 | 2.0 | 10.0 | 0.42 | 1.0 | 0.54 | 0.53 | 0.63 |

[a] effective density and effective temperature of the mixture, defined as $\rho\sigma_x^3$ and $kT/\varepsilon_x$ respectively, using Kay's rules for $\sigma_x$ and $\varepsilon_x$ (see Equation (7)).

**Table 2** Thermal conductivity of continuous mixtures with gaussian distributions.

| $\varepsilon_2/\varepsilon_1$ | $\sigma_2/\sigma_1$ | $M_2/M_1$ | $\rho^{*a}$ | $T^{*a}$ | $\lambda_0$ | $\lambda_2$ | $\lambda_1$ |
|---|---|---|---|---|---|---|---|
| Gaussian I | | | | | | | |
| 1.0 | 0.5 | 1.0 | 0.42 | 1.00 | 3.54 | 3.43 | 3.93 |
| 1.0 | 1.5 | 1.0 | 0.78 | 1.00 | 3.91 | 3.87 | 3.60 |
| 1.0 | 3.0 | 1.0 | 0.80 | 1.00 | 2.01 | 1.93 | 1.57 |
| 4.0 | 2.0 | 10.0 | 0.84 | 1.00 | 2.29 | 2.24 | 2.22 |
| 1.0 | 1.0 | 10.0 | 0.84 | 1.00 | 2.96 | 2.90 | 3.14 |
| 4.0 | 1.0 | 1.0 | 0.84 | 1.00 | 11.12 | 10.79 | 11.79 |
| 4.0 | 2.0 | 10.0 | 0.42 | 1.00 | 0.52 | 0.52 | 0.59 |
| Gaussian II | | | | | | | |
| 1.0 | 0.5 | 1.0 | 0.42 | 1.00 | 3.97 | 3.77 | 4.09 |
| 1.0 | 3.0 | 1.0 | 0.80 | 1.00 | 1.73 | 1.66 | 1.56 |
| 4.0 | 2.0 | 10.0 | 0.84 | 1.00 | 2.27 | 2.18 | 2.22 |
| 1.0 | 1.0 | 10.0 | 0.84 | 1.00 | 3.06 | 3.15 | 3.12 |

[a] effective values, see Table 1.

component characterization. The results show that the 2-component characterization gives results to within 5 percent of the continuous mixture value in all cases. The 1-component characterization, on the other hand, leads to considerable errors at the larger ratios (e.g., 33 percent for $\sigma_2/\sigma_1 = 3.0$ and 20 percent for $M_2/M_1 = 10.0$). When all three parameters vary simultaneously (roughly as in $C_1/C_{16}$ mixtures), it appears, that at high densities, some cancellation of errors takes place, and the 1-component characterization does better. However, this cancellation is somewhat erratic, as can be seen for the low density case ($\rho^* = 0.42$), and cannot in general be relied upon.

The results for continuous mixtures with Gaussian I and Gaussian II distributions are shown in Table 2. As was the case with linear distributions, the 2-component characterization gives results to within 5 percent of the continuous mixture value. The 1-component does better than in the linear distribution case and improves as the standard deviation decreases from Gaussian I to Gausian II. This could have been expected since there are more molecules with sizes closer to the average values in these cases. Here, as was the case with linear distributions, there is some cancellation of errors when all three parameters are varied, but leads to considerable errors at low densities.

The distributions discussed so far have all been symmetric (about $x_{\text{cumulative}} = 0.5$). Figure 3 shows two unsymmetric distributions studied by us as described by Equation (9). Unsymmetric I has a standard deviation of $(\sigma_2 - \sigma_1)/4$ and a mean of $(3\sigma_1 + \sigma_2)/4$. Unsymmetric II has the same standard deviation, but a mean of $(\sigma_1 + 3\sigma_2)/4$. The important difference as can be seen from Figure 3 is that Unsymmetric I has a larger number of smaller molecules while Unsymmetric II has a larger number of larger molecules. Two types of characterizations were studied to determine an accurate representation of such continuous mixtures. One is the usual $n$-component *equimolar* characterization used previously. The other, is an equiparameter characterization, which usually leads to nonequimolar mixtures. A 4-component equi-sigma characterization is shown in Figure 3 for Unsymmetric I, to further clarify such characterizations. In this case, all components within $\sigma_1$ and $(3\sigma_1 + \sigma_2)/4$ are characterized as one component, between $(3\sigma_1 + \sigma_2)/4$ and $(\sigma_1 + \sigma_2)/2$ as the second component and so on.

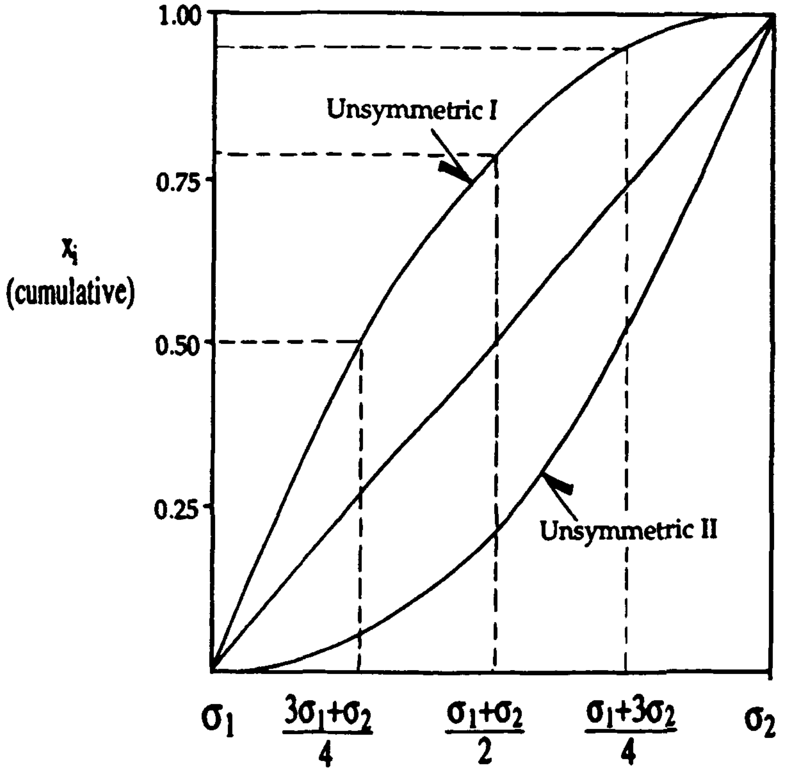

**Figure 3** Unsymmetric distributions for continuous mixtures. An example of a 4-component equi-sigma characterization is shown for Unsymmetric I. Here all components within $\sigma_1$ and $(3\sigma_1 + \sigma_2)/4$ are characterized as one component, between $(3\sigma_1 + \sigma_2)/4$ and $(\sigma_1 + \sigma_2)/2$ as the second component and so on.

The results for unsymmetric mixtures are shown in Table 3. As for all other distributions, the 2-component equimolar characterization gives satisfactory results for both Unsymmetric I and II. The equiparameter (equi-sigma, since the other ratios are 1.0), however, leads to larger errors. For example, the 2-component characterization is 11 percent off (compared to $\lambda_0$) for the Unsymmetric II distribution for the size

**Table 3** Thermal conductivity of continuous mixtures with unsymmetric distributions.

| $\varepsilon_2/\varepsilon_1$ | $\sigma_2/\sigma_1$ | $M_2/M_1$ | $\rho^*$[a] | $T^*$[a] | $\lambda_0$ | $\lambda_2$ | $\lambda_1$ |
|---|---|---|---|---|---|---|---|
| Unsymmetric I | | | | | | | |
| 1.0 | 1.5 | 1.0 | 0.78 | 1.00 | 4.61 | 4.52[b]<br>4.43[c] | 4.29 |
| 1.0 | 3.0 | 1.0 | 0.80 | 1.00 | 2.97 | 2.85[b]<br>2.76[c] | 2.32 |
| Unsymmetric II | | | | | | | |
| 1.0 | 3.0 | 1.0 | 0.80 | 1.00 | 1.38 | 1.32[b]<br>1.23[c] | 1.09 |

[a] effective values, see Table 1.
[b] equimolar characterization.
[c] equiparameter characterization.

**Table 4** Thermal conductivity of semi-continuous mixtures.

| $\varepsilon_2/\varepsilon_l$ | $\sigma_2/\sigma_l$ | $M_2/M_l$ | $\rho^{*a}$ | $T^{*a}$ | $\lambda_0^b$ | $\lambda_2$ | $\lambda_l$ |
|---|---|---|---|---|---|---|---|
| 1.0 (A) | 3.0 | 1.0 | 0.80 | 1.00 | 2.50 | 2.55 | 1.80 |
| 1.0 (B) | 3.0 | 1.0 | 0.80 | 1.00 | 2.23 | 2.19 | 1.56 |
| 1.0 (C) | 3.0 | 1.0 | 0.80 | 1.00 | 1.97 | 1.87 | 1.42 |
| 1.0 (D) | 3.0 | 1.0 | 0.80 | 1.00 | 1.66 | 1.58 | 1.28 |
| 1.0 (E) | 3.0 | 1.0 | 0.80 | 1.00 | 1.68 | 1.61 | 1.24 |

[a] effective values, see Table 1.

[b] $\lambda_0$ represents the actual thermal conductivity of the semi-continuous mixture.

(A), (B), (C), (D), (E) are five different semi-continuous mixtures. See text for details.

ratio of 3.0. Of course, as the number of components increase, both equiparameter and equimolar would ultimately give the same results. We found that with equi-sigma, one needs to go to at least 4 components to obtain satisfactory results. It was unnecessary to investigate equiparameter characterizations for gaussian distributions, since for all symmetric distributions equiparameter and equimolar are identical for 2-component characterizations.

Several semi-continuous mixtures were also examined. Mixtures with symmetric and unsymmetric distributions and containing upto 4 discrete components were included in this study and were modelled as before with $n$ equimolar components. Here the continuous mixture thermal conductivity was obtained by modelling the continuous part as 2 equimolar components (since our results show that the 2-component equimolar characterization of a continuous mixture is satisfactory) while the discrete components were kept unchanged. Table 4 shows the results obtained for five different semi-continuous mixtures labelled (A) through (E). (A) has two discrete components with sizes $1.25\sigma_1$ and $1.50\sigma_1$ and mole fractions of 0.2 and 0.3 respectively. The continuous part has an unsymmetric distribution (type I of Figure 3) between $2.0\sigma_1$ and $3.0\sigma_1$. (B), (C) and (D) all have one discrete component and a gaussian distribution (type I of Figure 1) between $2.0\sigma_1$ and $3.0\sigma_1$. The discrete component has a size of $1.50\sigma_1$ but the mole fraction is 0.5, 0.4 and 0.25 for (B), (C) and (D) respectively. Finally (E) has four discrete components with sizes $1.25\sigma_1$, $1.40\sigma_1$, $1.50\sigma_1$ and $1.90\sigma_1$ with mole fractions 0.15, 0.03, 0.02 and 0.05, respectively. The continuous part has a gaussian distribution (type I of Figure 1) between $2.0\sigma_1$ and $3.0\sigma_1$. We see that the 2-component equimolar characterization does an adequate job of characterizing the continuous mixture for all the semi-continuous mixtures studied.

**Table 5** Thermal conductivity of discrete mixtures for $\rho^* = 0.80$, $T^* = 1.00$, $M_2/M_1 = 1.0$ and $\varepsilon_2/\varepsilon_1 = 1.0$.

| *Mixture* | *molefraction* | $\sigma$ | $\lambda_0^a$ | $\lambda_2$ | $\lambda_l$ |
|---|---|---|---|---|---|
| (A) | 0.2<br>0.4<br>0.2<br>0.2 | 1.0<br>1.2<br>1.4<br>1.6 | 4.30 | 4.09 | 3.83 |
| (B) | 0.2<br>0.2<br>0.2<br>0.4 | 1.0<br>1.2<br>1.4<br>1.6 | 3.95 | 3.79 | 3.48 |

[a] Here $\lambda_0$ is the actual thermal conductivity of the 4-component mixture.

Table 5 shows two discrete mixtures that were modelled as 1- and 2-component equimolar mixtures and compares them with the actual discrete mixture values. In both cases the 2-component gives results close to the discrete mixture thermal conductivity. The 1-component underpredicts by upto 12 percent.

## CONCLUSIONS

The thermal conductivity of continuous, semi-continuous, and discrete mixtures has been evaluated. Various distributions have been examined including, linear, gaussian and unsymmetric distributions. We showed that the 2-component equimolar characterization gives adequate results for a wide range and types of distributions. In addition, it is not necessary to go to higher order characterizations since this does not improve results any further. We also showed that equiparameter characterization of mixtures with unsymmetric distributions can lead to larger errors. Our 1- and 2-component characterizations are similar in spirit to the 1- and 2-fluid theories sometimes used in thermodynamics. However, since we are using molecular dynamics, we did not have to assume that the two components mix ideally - an assumption made in the 2-fluid theory. It appears from our results that a good theory for the thermal conductivity of 2-component mixtures can perhaps be used to predict the thermal conductivity of many component mixtures.

### *Acknowledgements*

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## APPENDIX: DETAILS OF THE NEMD SIMULATION

Here we outline the details of the simulation technique employed. The simulations were carried out on 256 molecules using a spherical cutoff of $r_c^* = 3.0$. The initial configuration was that of an FCC. Configurations from a previous simulation were used when possible. Equilibration took 5000 to 10,000 time steps and an additional 50,000 to 70,000 time steps were needed for the thermal conductivity to converge. We used a reduced time step of 0.002. The two first order equations (Equations (1) and (2)) were solved using Gear's fifth order predictor-corrector scheme. The range of external fields used in this work were $F(t)^* = 0.1, 0.2, 0.3$ in reduced units. We found that within statistical uncertainties there was no observable dependence of the conductivity with respect to the magnitude of the external field. The gaussian thermostat ($\alpha$ in Equation (2)) was used to control the temperature. With the thermostat on and velocity rescaling done every time step the temperature was kept correct to five significant figures. Linear momentum was conserved. As a further check, we tested the program with the thermostat turned off and $F(t)^* = 0$ (conventional EMD) to ensure that energy conservation is obtained. The thermal conductivities obtained from these simulations were found to agree well with previously published results (from MD simulations) for both pure fluids and mixtures [1, 8].

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