Coarse-graining strategy for reducing multidispersity at fixed particle density: A dynamical study

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Abstract

We present a coarse-graining method aiming to reduce multidispersity in complex fluids using the example of the well-established bidisperse Lennard-Jones mixture discovered by Kob and Andersen. Our method foots on the iterative Boltzmann inversion scheme and is designed to conserve the total particle number. The preservation of the characteristic dynamics of this bidisperse mixture is the focus of our investigation. It turned out that it is indeed possible to find a one-particle model with similar dynamic features, like glass transition temperature, at the cost of structural similarity.

1 Introduction

In the following, we introduce a novel type of coarse-graining procedure for isotropic systems comprising more than one particle species. By coarse-graining (CG), we understand the systematic treatment of microscopic details and dynamics from a coarser perspective. Common methods in the literature include force matching schemes, the relative entropy method, the conditional reversible work method, the inverse Monte Carlo method, the
iterative Boltzmann inversion method\textsuperscript{9,10} or hybrid schemes.\textsuperscript{11} The main idea is to develop force fields among a small set of coordinates, called reaction coordinates\textsuperscript{12,13} or collective variables, that form projections of microscopic degrees of freedom. This has the computational advantage of evaluating a smaller set of equations of motion. However, CG has also the physical advantage that effective particles, like molecules, which are described via few collective variables (e.g., the molecules’ center-of-mass positions\textsuperscript{14}) and effectively comprise a large number of microscopic variables (e.g., atom positions), interact via smooth effective pair potentials which are not overlaid by time-dependent noise caused by atomic vibrations, i.e. the potential energy surface becomes very smooth. This smooth energy landscape leads to a faster particle diffusion (see Ref. \textsuperscript{15}), and accordingly to shorter relaxation times within the coarse-grained system. The latter fact also tends to result in a computational advantage when calculating equilibrium quantities.

Our here presented coarse-graining method differs from the previously described standard strategy since we aim at effectively reducing the number of species while not reducing the particle density. By forcing such constraint, we satisfy that the effective system becomes indistinguishable from the original system when approaching the high-temperature limit. This is based on the fact that with rising temperature the pair potentials become flatter in units of thermal energy (which is proportional to $T$). Another fundamental demand within our CG procedure is that the all-particle radial distribution function (RDF), which does not distinguish between the particle type, is set to remain unchanged when reducing the multidispersity. To test our method, we chose the Kob-Andersen (KA) mixture\textsuperscript{16,17}, which is a bidisperse Lennard-Jones mixture whose range and strength parameters do not fulfill the common Lorentz-Berthelot mixing rule.\textsuperscript{18} The KA mixture, when supercooled but still being warmer than its glass transition temperature, has a very long lasting isotropic regime before it crystallizes. Mixtures with similar dynamic features also exist for other models,\textsuperscript{19,20} even in different dimensions.\textsuperscript{21,22} By creating a coarse-grained or effective KA system consisting of a one-particle species as schematically shown in Fig. 1, a faster diffusion
and relaxation time might result, which significantly reduces the computational cost for obtaining equilibrium quantities. As the principal method for developing an effective pair potential yielding the same RDF we chose the iterative Boltzmann inversion (IBI) method.\textsuperscript{[9]} In contrast to other CG approaches aiming at conserving dynamic features,\textsuperscript{[23,24]} the IBI method conserves with the RDF an equilibrium quantity. Thus, it is interesting to scrutinize if, or under which conditions, the here presented CG method is still able to maintain the ability of the system to appear in a glass phase. Another interesting aspect of this CG procedure concerns the stability of the supercooled phase since monodisperse systems tend to have shorter relaxation times and crystallize at a fast pace. Prior approaches for preventing crystallization in monodisperse model fluids include shape anisotropy,\textsuperscript{[25]} pinned particles,\textsuperscript{[26]} ultrasoft interactions\textsuperscript{[27]} or the use of a double well\textsuperscript{[28]} as well as an undulating pair potential.\textsuperscript{[29]} As we later see, our approach also leads to undulating pair potentials.

The remainder of this article is organized as follows. In Sec. 2 we present the theory describing the transformation from the bidisperse system to the effective monodisperse system. Then in Sec. 3 we investigate static structural properties as well as dynamic aspects of our effective system. Finally, we conclude our findings in Sec. 4.

2 Model and coarse-graining procedure

In this section, we present our CG procedure describing a transition from a bidisperse many-particle system towards a monodisperse one using the example of the supercooled Kob-

Figure 1: Transition from the bidisperse Kob-Andersen fluid (particle species A=red, B=turquoise) towards an effective monodisperse fluid (yellow particles).
Andersen (KA) mixture. The latter is a mixture of Lennard-Jones particles of mass $m_p$ comprising two different species, labeled with A and B, whereas there are four times more A than B particles. It is these few B particles which prevent the other particles, that form the 80% majority, from quickly crystallizing. The set of pair potentials as a function of the inter-particle distance $R$ in that mixture is defined through

$$U_{\alpha\beta}(R) = \begin{cases} U_{LJ}(R, \epsilon_{\alpha\beta}, \sigma_{\alpha\beta}) - U_{LJ}(2.5\sigma_{\alpha\beta}, \epsilon_{\alpha\beta}, \sigma_{\alpha\beta}), & R \leq 2.5\sigma_{\alpha\beta} \\ 0, & \text{else} \end{cases}$$

(1)

with $\alpha, \beta \in \{A, B\}$, and the Lennard-Jones parameters for the well depth

$$\epsilon_{AA} = \epsilon, \quad \epsilon_{AB} = 1.5\epsilon, \quad \epsilon_{BB} = 0.5\epsilon$$

and contact distance

$$\sigma_{AA} = \sigma, \quad \sigma_{AB} = 0.8\sigma, \quad \sigma_{BB} = 0.88\sigma.$$ 

Corresponding potential curves are depicted in Fig. 2 as dashed lines. In accordance with the original KA system, we consider in the effective monodisperse system the same particle density $\rho$ being fixed at $\rho\sigma^3 = 1.2$. The fundamental idea behind our coarse-graining approach is to identify at each considered temperature the all-particle RDF (denoted with $g$) of the bidisperse KA with the RDF of the coarse-grained (CG) or effective monodisperse system, i.e.

$$g_{KA} \overset{!}{=} g_{CG}.$$ 

(2)
This is equivalent to identifying the mean force profiles. The definition of the all-particle RDF based on the $N$ particle positions $\{r_i\}$ that is

$$g(R) = \frac{2}{\rho \cdot (N-1) \cdot 4\pi \cdot R^2} \left\langle \sum_{i<j\in\{1,...,N\}} \delta(|r_j-r_i|-R) \right\rangle,$$

is designed to not distinguish among particle species. In the next step, we set out to find a monodisperse isotropic system with the same particle density, whose RDF is equivalent to the one in the KA system. Moreover, for computational reasons, it would be desired if the total potential energy in that effective system has only pair-wise contributions. The existence of such underlying pair potential, however is not proven, but as soon as a pair potential exists, it is unique according to Henderson. We further want to point out that such a pair potential not only depends on temperature but also on the particle density. More specifically, if we choose another particle density, but same temperature in the KA system, we will obtain a different effective pair potential. For the interested reader we recommend the work of A. A. Louis thematizing a discussion concerning density-dependent pair potentials.

In order to obtain the effective pair potential, we chose the iterative Boltzmann inversion (IBI) method. The corresponding procedure requires that we simulate at each iteration a monodisperse system with an approximate pair potential $u_i$ and determine an improved pair potential...
potential $u_{i+1}$ by making use of the obtained RDF $g_i$ according to the following iterative formula

$$u_{i+1}(R) = u_i(R) - \alpha_i(R) \ln \left( \frac{g_i(R)}{g_{KA}(R)} \right).$$  \hspace{1cm} (4)

As an initial pair potential, we choose the solution of the hypernetted chain approximation (HNC),\textsuperscript{33} that is

$$u_0(R) = u_{HNC}(R) = -k_B T \ln(g_{KA}(R)) + k_B T \cdot (g_{KA}(R) - c_{KA}(R) - 1),$$  \hspace{1cm} (5)

with $c_{KA}$ being the direct correlation function (implicitly defined through the Ornstein-Zernike equation\textsuperscript{34} but explicitly defined in k-space\textsuperscript{33}) of the KA system. If the functions $\alpha_i(R)$ in Eq. (4) are carefully chosen, we can reach convergence after a certain iteration index $j$, i.e. $g_{i\geq j} \approx g_{KA}$. For this purpose, we implemented in accordance with the original literature\textsuperscript{9} the following mixing function

$$\alpha_i(R) = \alpha_0 \cdot k_B T \cdot \exp(-R^2/(2 \kappa^2))$$  \hspace{1cm} (6)

with $k_B$ being the Boltzmann constant and the remaining parameters were set to $\alpha_0 = 0.05$ and $\kappa = 4\sigma$. The latter parameters are fixed for all $i$ and represent the main mixing parameter as well as the effective range. The overall particle number in all systems was set to 35000, which is rather large compared to the original work of Kob and Andersen from 1994,\textsuperscript{16} but allows on the one hand to correctly track the generally long-range character of the RDFs $g_i$ and avoids system size effects on the other. With respect to the pair forces, a cut-off distance of $5\sigma$ turned out being sufficient. All simulations for each iteration were performed with the Gromacs simulation package 4.6.7 covering 60000 time steps per iteration, whereas a time step has a length of 0.005 time units (t.u.) being $\sigma \cdot (m_p/\epsilon)^{1/2}$, i.e. each iteration covers a time interval of 300 t.u. In order to obtain the reference RDF $g_{KA}$ at each considered temperature,
we simulated each bidisperse KA system with even half the time step length and extracted the required positional data from the time interval $[7000 \, \text{t.u.}, \, 15000 \, \text{t.u.}]$. As initial systems for the bidisperse as well as all initial monodisperse systems, we used random configurations that were quickly equilibrated using the steepest descent method, and in order to not disturb the systems at each iteration too much, we additionally took the last system snapshot of the $i$-th iteration as input for iteration $i + 1$. Considering the temperature control, we used the Nosé-Hoover thermostat with a time constant of 1 t.u.

After about 1200 iterations, convergence has been achieved in all our systems in the considered (dimensionless) temperature range covering $T^* = k_B T / \epsilon = 0.025, 0.05, 0.1, 0.15, \ldots, 1$. At higher temperatures the system is above the freezing point and accordingly out of supercooling. The total iteration number of 1200 seems to be quite large but its high value stems from the low choice of $\alpha_0$. The RDFs $g_i$ are obtained using the last third of the positional data of each iteration run. Though, this only covers 100 t.u., it leads to the convergence of the IBI scheme as exemplary shown in Fig. 3 for the $g_i$ and $u_i$ at temperatures being with (a) $T^* = 0.6$ above and with (b) $T^* = 0.4$ below the glass transition temperature ($T_g \approx 0.435$). At both temperatures, the first iteration of the RDF, $g_1$ (being the RDF corresponding to $u_{HNC}$) in Figs. 3(a-b) reveals a low-valued first peak (at $R \approx 0.85 \sigma$) and a high-valued second peak (at $R \approx 1.05 \sigma$) with respect to the specific reference RDF $g_{KA}$ from the bidisperse KA system. In spite of this observed similar shape characteristics for both $g_1$ at these two temperatures, we detect during the first 400 iterations a significant difference in the convergence behavior that can be observed by analyzing the corresponding corridor of solutions marked in red. At $T^* = 0.6$ (Fig. 3(a)) that corridor and the series of corridors reveals a good-natured convergence, which is reflected by their location between $g_1$ and $g_{1200}$, whereas at $T^* = 0.4$ (Fig. 3(b)) the corridor covering the first 400 iterations is quite broad and shows a strong tendency to over- and underestimate peak heights. Such an extreme overestimation of the first peak is observed since the system shows cavitation effects driven by a negative pressure (see snapshot in Fig. 3(c)). However, these effects become less
Figure 3: Iterative Boltzmann inversion (IBI) results covering 1200 iterations in a 35000 particle Kob-Andersen system for the radial distribution functions (RDFs) $g_i$ (defined through Eq. (3)) at (a) $T^* = 0.6$, (b) $T^* = 0.4$ and the corresponding pair potentials $u_i$ at (c) $T^* = 0.6$, (d) $T^* = 0.4$ are displayed through colored regions. Curves in (a, b): the first IBI iteration for the RDF $g_1$ (dashed line); the reference RDF $g_{KA}$ (solid line); final RDF after 1200 steps (green dots). Figure (b) additionally contains a snapshot of our system temporarily collapsing at IBI-step 213. Curves in (c, d): initial potential $u_{HNC}$ according to Eq. (5) (dashed line); final result for the effective potential after 1200 iterations (solid line).
pronounced the more steps we took into account, and as we later see, the converged RDFs all stem from isotropic phases in which the system can not overcome a critical cavitation within reasonable simulation time and no crystallization is present. Despite the observed stability, the pressure is in part also negative for our converged simulations as can be seen in Fig. 4. In that figure the pressure progression along the temperature is shown for the bidisperse and the effective monodisperse system through a black-solid and a thick black-dashed line, respectively. We recognize that the effective monodisperse system reveals a far lower pressure than the bidisperse system as a consequence of the altered packing fraction. In particular, the particle radius in the effective monodisperse system, as defined by the intersection of the potential with zero, is similar to the A-B configuration (see Fig. 2 showing the Kob-Andersen pair potentials and the effective pair potentials from the IBI analysis displayed in Fig. 3) marking the closest possible combination among particle pairs. From this size definition it

![Pressure as a function of temperature](image)

Figure 4: Pressure as a function of temperature covering the original bidisperse Kob-Andersen system, the effective monodisperse system and the later introduced non-intrinsic temperature dependent monodisperse models M0.45 and M0.9 evaluated using the long sampling scheme (standard) and additionally the short sampling scheme. A horizontal daled line marks the pressure=0 line.
follows that the effective system has a lower packing fraction. Effective particles thus can get very close before feeling core repulsion which would significantly increase the pressure in a close packed system.

Coming back to our IBI convergence analysis, we recognize that in contrast to the initially alternating convergence behavior of the $g_i$ functions at low temperatures, the associated pair potentials $u_i$ show a rather well-behaved convergence at all temperatures as exemplarily shown in Figs. 3(c-d). This is a consequence of our low-valued mixing parameter $\alpha_0$. Even though the value of $\alpha_0$ appears small, it provides a feedback that is strong enough to prevent even supercooled systems close to the glass transition from quickly crystallizing. By scrutinizing the deviation between the converged pair potential $u_{1200}$, and the initial HNC pair potential $u_{\text{HNC}}$ in Figs. 3(c-d), we detect a strong alignment at large inter-particle distances ($R \gg \sigma$) which is a known feature of the HNC approximation. However, the HNC approximation failed to correctly predict the effective pair potential $u$ at small inter-particle distances $R$. Therefore, the IBI scheme—or perhaps some promising alternative—turned out being necessary, even though its implementation and convergence in a large simulation system is involved with respect to computational requirements. We further would like to point out that the values for $u_i$ below $R = 0.8\sigma$ were extrapolated by cumulatively integrating the pair force along $R$, which we have assumed being equal to the force between A and B particles in that close-contact regime.

All in all, our IBI convergence behavior seems quite weak compared to the original work of Soper, who used IBI to simplify water models with a mixing parameter of $\alpha_0 = 1$. Our system, however, is considered in the supercooled phase, which is rather unstable at temperatures slightly above the glass transition temperature. Even though the system below the glass transition temperature is quite stable for a very long time, approximate solutions during the IBI scheme might not (see Fig. 3(b)). Another problematic issue for reaching convergence in the IBI scheme is the high particle density increasing the likelihood of clumping due to a higher number of next neighbors.
3 Structural and dynamical analysis

In the current section, we present an analysis of the equilibrium structure and dynamics of the bidisperse and the effective monodisperse KA system at various temperatures. Regarding the curves of the converged pair potentials \( u_{1200} \) from the IBI scheme shown in previous section (Figs. 3(c-d)), we can identify an alternating behavior along the inter-particle distance \( R \); especially a strong repulsive shoulder appearing at around \( R = 1.5\sigma \). To have a comparison of this effective pair potential with pair potentials of the original KA system, we have included \( u_{1200} \) from Fig. 3(c) already in Fig. 2.

Our next objective is the investigation of the influence of temperature on the reference RDF \( g_{KA} \) and its associated converged pair potential \( u_{1200} \). Corresponding plots are depicted in Fig. 5. The investigated temperatures were chosen being above the glass transition temperature or critical temperature from the mode-coupling theory (MCT) of the bidisperse KA system. For this purpose, the MCT predicted a value of \( T^* = 0.435 \). Results at lower temperatures, at which the system’s relaxation time becomes quite large (even infinite in MCT) and ensemble averages generally sprawl over large time intervals, are represented with blue curves. As expected, we observe in the RDFs (Fig. 5(a)) a slight progression towards the value 1 (ideal gas limit) the higher the temperature. But as depicted in the inset, this progression seems to slow down at high temperatures since the system resembles with its steep potentials a hard-sphere mixture before also these pair potentials become softer at higher temperatures. The associated pair potentials (Fig. 5(b)), however, still reveal a high change rate among neighboring temperature sets. This is due to the fact that similar RDFs imply a similar Boltzmann weight factor leading to a nearly direct proportionality between \( u \) and \( T \) in the canonical ensemble average for overcritical temperatures (see inset). This behavior is in contrast to the temperature dependence of effective pair potentials of particles whose internal degrees of freedom were coarse-grained. As a result, the pair potential in units of \( k_B T \) (Subfig. (c)) becomes only slightly weaker with increasing temperature. Only values at \( R \approx 0.9\sigma \) show stronger weakening effects alongside the temperature progression.
We interpret it as a consequence of temperature-driven combinatorically higher likelihood of the A-A particle interaction with its Lennard-Jones well depth at $R \approx 1.1\sigma$.

We now turn the focus towards dynamic properties to assess the changes when transforming from the bidisperse to the effective monodisperse system. In this regard, we next investigate the mean squared displacement defined through

$$\text{MSD}(t) = \frac{1}{N} \left\langle \sum_i |r_i(t) - r_i(0)|^2 \right\rangle$$

for the bidisperse and the effective monodisperse system in Fig. 6(a)-(b) for the temperature range $T^* = 0.7, \ldots, 0.45$ and $T^* = 0.7, \ldots, 0.1$, respectively. Corresponding ensemble averages (indicated by \langle \ldots \rangle) were taken in the time interval $[7000 \, \text{t.u.}, 30000 \, \text{t.u.}]$. We, from now on, refer to an evenly sampling (as used to approximate the ensemble average) within this long time interval as long sampling scheme (forming the standard sampling interval within our investigation). As expected, we detect in the bidisperse system (Fig. 6(a)) a pronounced appearance of a subdiffusive regime, where the MSD is almost approaching a constant value when cooling the system. This is reflected by the plateau separating ballistic motion characterized by $\text{MSD}(t) \propto t^2$, and diffusive motion, at which $\text{MSD}(t) = 6Dt$ holds, with $D$ being the diffusion coefficient. Within the same temperature range in the effective monodisperse system (Fig. 6(b)), no such plateau emerges, i.e. the ballistic regime is directly followed by the diffusive regime. However, by further cooling the system a slightly subdiffusive regime emerges which is still revealing very high MSD values.

In an effort to overcome this drawback, we propose in the following a simpler model, for which later turned out, it has better consistency in the dynamics at the cost of having structural equivalence. Our new choice consists in using for all temperatures the effective monodisperse model developed at a specific temperature $T_{\text{ref}}$ and denote this model as $MT^*_{\text{ref}}$ with $T^*_{\text{ref}} = k_B T_{\text{ref}}/\epsilon$. All new simulations from that model were started from an isotropic configuration for which we took the equilibrated configuration of the effective monodisperse
Figure 5: Results for (a) the RDF in a 35000 particle Kob-Andersen system at different temperatures (in dimensionless form: $T^* = k_B T/\epsilon$) and corresponding pair potentials defined through Eq. (4) with $i = 1200$ in units of (b) $\epsilon$ and (c) $k_B T$. 
Figure 6: Mean squared displacement for a broad range of temperatures (in dimensionless form: $T^* = k_B T/\epsilon$) for (a) the original Kob-Andersen system, (b) the effective monodisperse system and (c) the monodisperse system whose pair interaction is given through the non-intrinsic temperature dependent M0.9 model. Subfig. (c) additionally contains results for a short sampling scheme (for details see main text). The circle marks curves of interest.
system at $T_{\text{ref}}$, but gave it enough time to relax depending on the equilibrium sampling scheme used for the determination of static as well as dynamic quantities. This model has by definition no intrinsic temperature dependence. The idea behind this model is to lower the high diffusion (seen in Fig. 6(b)) by avoiding the significant smoothing effect of the effective pair potential towards low temperatures as depicted in Fig. 5(b). By having a look at the M0.9 model—which as we later see shows a similar glass transition temperature as the reference KA system—we detect for the MSD curve at $T^*=0.45$ (as depicted in Fig. 6(c) with solid lines) a pronounced shape conformity with the one of the reference system (cp. Fig. 6(a) with Fig. 6(c)). However, the temperature progression of the MSD curves of model M0.9 reveals some abrupt change between 0.55 and 0.5. By inspecting system snapshots, we found that the systems crystallize at $0.435 \lesssim T^* \lesssim 0.5$ while we denote a faster pace at $T^*=0.5$ compared to $T^*=0.45$ within the framework of our simulations. This explains that the MSD at $T^*=0.5$ is even lower than at $T^*=0.45$. However, at lower temperatures no signs of crystallization have been detected which might indicate the glassy state. From the literature\cite{37} we know that the bidisperse KA system crystallizes at a very long time scale since the one particle species prevents the other species from crystallizing. So these species even segregate before crystallizing.\cite{37} Segregation takes a lot of time and is not happening in a monodisperse system by definition. In order to cope with this crystallization issue, we determined the MSD also in a shorter time interval right after starting the simulation. In particular, we hereby took ensemble averages in this so-called short sampling scheme covering the time interval $[100\; \text{t.u.}, 1100\; \text{t.u.}]$. Corresponding results are depicted in Fig. 6(c) with dashed curves, which coincide with prior results (solid curves) at temperatures slightly higher than $0.5\epsilon/k_B$, and significantly diverge for temperatures slightly below, at which the system crystallizes. However, at temperatures below the glass transition temperature, the system will never crystallize (when assuming the thermodynamic limit). In accordance with the other models, we have also investigated the pressure for our M0.9 model and additionally the M0.45 model in the prior introduced Fig. 4, which are represented by orange and purple
lines, respectively. As expected, we recognize also very low pressures in both of these non-intrinsic temperature dependent modeled systems compared to the bidisperse one. With respect to the M0.9 modeled system no negative pressure for temperatures $T^* \gtrsim 0.3$ has been detected. We therefore observe for this system that the glass transition temperature—which can be estimated from the MSD analysis (Fig. 6(c))—is already in the temperature range with positive pressure such that cavitation is not appearing there. Contrary to the short sampling scheme we observe in the long sampling scheme, close but above the glass transition temperature, that the pressure curve reveals for both models a kink which peaks at about the highest temperature where crystallization has been observed. At that temperature(s) the dynamics is fast enough to establish a phase transition within the long sampling scheme and cool enough to account for crystallization.

In order to check whether there is an overall isotropy or a crystallization in the effective systems, we next analyze the particle projections onto the x-y, y-z and z-x plane and investigate the corresponding two-dimensional RDFs. These are defined as

$$g_{xy}(w) = g_{\perp}(w, (0, 0, 1)),$$

$$g_{yz}(w) = g_{\perp}(w, (1, 0, 0)),$$

$$g_{zx}(w) = g_{\perp}(w, (0, 1, 0)),$$

$$g_{\perp}(w, \hat{n}) = \frac{1}{(N - 1) \cdot (N \rho^2)^{1/3} \cdot \pi w} \left\langle \sum_{j,k>j} \delta\left(w - |\hat{n} \times R_{jk}|\right) \right\rangle,$$

with $\hat{n}$ being the normal vector for a considered plane. In Fig. 7(a), we present resulting curves of Eqs. (8a)–(8c) for the bidisperse system at the temperatures $T^* = 0.3, 0.45, 0.5, 0.55, 0.7$. From these 2D-RDF curves we detect that there is indeed no dependence on the choice of orthogonal planes, i.e. there is no global structuring in the bidisperse system as expected. Also, the temperature influence is weak, which is in accordance with the small temperature dependence of $g_{KA}$ depicted in Fig. 5(a) in the same temperature range. These characteristics also holds in the effective monodisperse system as depicted in Fig. 7(b). That means
Figure 7: Distribution functions defined by Eqs. (8a)–(8d) at various temperatures (in dimensionless form: $T^* = k_B T/\epsilon$) covering the range from $T^* = 0.7$ with small orange dots to $T^* = 0.3$ with large circled black dots (alternatively lines for $T^* = 0.45, 0.5, 0.55$) for (a) the bidisperse Kob-Andersen system, (b) the effective monodisperse system and for the M0.9-modeled system for (c) the long, and (d) short sampling scheme (for details see main text).
both, the bidisperse and the effective monodisperse systems’ structure is isotropic and there is no sign of an abrupt structural change with temperature which would indicate a phase transition. Moreover, all the curves are quite alike. With respect to model M0.9, we expect at least some change since the underlying pair potential is no longer a function of temperature. Corresponding distribution functions are shown in Figs. 7(c-d) and reveal that there is in fact abrupt structural change at larger time scales when scrutinizing the progression along the temperature, especially at small 2D inter-particle distances. Subfig. (c), which depicts all three distribution functions at a larger time scale (long sampling scheme used) displays the anisotropy resulting from the crystallization by having significantly different values at \( w \lesssim 3\sigma \) at low temperatures \( (T^\ast = 0.45, 0.5) \), whereas the system remains purely isotropic at shorter time scales (short sampling scheme used) as depicted in Subfig. (d). For the latter case, no significant structural change can be detected, neither as a function of temperature nor with respect to the plane in which we evaluate \( g_\perp \). The structural change in Subfig. (c) is even higher at \( T^\ast = 0.5 \) than at \( T^\ast = 0.45 \), which is an artifact stemming from not enough equilibration time given in this slowly crystallizing system, and as we see in the forthcoming investigation, reflects that this model is close to its glass transition temperature.

In order to assess the glass transition temperature region, we present in Fig. 8 results for the diffusion coefficient \( D \) (definition in the caption) as a function of temperature \( T \) for the bidisperse and effective monodisperse model as well as the M0.45 and M0.9 model. We further provide for the M0.45 and M0.9 models results for both equilibrium sampling schemes since we have observed the mentioned crystallization phenomena. From this diagram, we can see that \( D \) in the bidisperse system follows a path with a monotonous positive slope while having vanishing values below the critical temperature. The curve of the effective monodisperse system follows also a positive trend but shows no inflection point throughout all temperatures. Consequently, a glass phase cannot be realized. The \( MT^\ast_{\text{ref}} \) models on contrary reveal an inflection point that can be shifted depending on the choice of \( T^\ast_{\text{ref}} \). By closer inspecting their glass transition region (inset of Fig. 8), we denote for \( D \lesssim 2 \cdot 10^{-3} \times \sigma \).
Figure 8: Diffusion coefficient defined by $D = \lim_{t \to \infty} \frac{MSD(t)}{6/t}$ for the bidisperse Kob-Andersen system (black-solid), the effective monodisperse system (black-dashed) and for the M0.45 and M0.9 modeled systems. Curves resulting from the short sampling scheme are added for the latter models (colored-dashed). The inset magnifies the transition regions between glass and supercooled liquid. Additional simulations in that magnified region have been performed to satisfy a $0.1 \epsilon/k_B$ temperature resolution.
\((\epsilon/m_p)^{1/2}\) a sudden decay of the diffusion, whereas this is not observed when giving the system less time to evolve by applying the short sampling scheme (dashed line). This phenomenon is consistent with our prior observation of the unusual progression of the MSD at the same temperatures (see Fig. 6(c)) due to the crystallization leading to positional restraints and accordingly vanishing MSD or diffusion.

We next scrutinize the relaxation of the systems in anticipation of an unusual behavior in the temperature regime, at which the MSD forms a plateau or shows a significant sub-diffusive behavior. For this purpose, we present in Fig. 9 results for the self-intermediate scattering function, that is

\[ F_s(k, t) = \frac{1}{N} \sum_i \cos(k |r_i(t) - r_i(0)|). \]

This function characterizes the relaxation of a density mode with wave vector \(k\). We hereby focus on the spherical component and further set the wavenumber \(k\) equal to the one corresponding to the maximum of the structure factor belonging to the RDF at \(T^* = 0.45\), yielding \(k\sigma \approx 7.3\). As expected, the bidisperse system (Fig. 9(a)) reveals the characteristic two-step relaxation behavior, which was already observed for each particle species separately. The effective monodisperse system (Fig. 9(b)), on contrary, is showing no traces of such behavior when lowering the temperature. This result is no surprise given the fact that the MSD showed no significant subdiffusive behavior (as shown in Fig. 6(c)) in which a first relaxation process would take place. Fortunately, a two-step relaxation dynamics is recovered within our \(MT^*\) models as exemplarily shown for M0.9 in Subfig. (c). In coincidence with the MSD, we also detect slightly below \(T^* \lesssim 0.5\) a strong influence of equilibrium scheme on \(F_s\). In fact, there is no more alignment between curves stemming from different equilibrium sampling schemes. The long sampling scheme in that temperature region even causes \(F_s\) to show an irregular pattern along the temperature. In particular, \(F_s\) at \(T^* = 0.5\) possesses higher values in the \(\alpha\)-relaxation regime compared to \(T^* = 0.45\), due to a crystallization
Figure 9: Self-intermediate scattering function defined through Eq. (9) for a broad range of temperatures (in dimensionless form $T^* = k_B T/\epsilon$) for (a) the Kob-Andersen system, (b) the effective monodisperse system and (c) the M0.9-modeled system covering 35000 particles. Subfig. (c) contains also results for the short sampling scheme (dashed curves). The circle marks curves of interest. Intersections of the curves with $1/e$ define the $\alpha$-relaxation times.
whose creation process is already finished within the long sampling scheme at $T^* = 0.5$ but not finished at $T^* = 0.45$. The short sampling scheme, however, leads to a rather “normal” temperature progression in that temperature regime (see next paragraph for more details), since the system is not given enough time to crystallize such that it remains supercooled as the bidisperse system.

In order to analyze the temperature progression of $F_s$ qualitatively, we next focus on its decay behavior and thereby quantify the $\alpha$-relaxation regime. Additionally, this also leads us to the characterization of the type of glass former by focusing on the alpha relaxation time $\tau_\alpha$ being implicitly defined through

$$F_S(\tau_\alpha) = 1/e. \tag{10}$$

By determining the $\tau_\alpha$ values at different temperatures for each model, we can see in Fig. 10(a) that in the bidisperse system, as well as the $MT^*_\text{ref}$ modeled systems, the following relation

$$\tau_\alpha \propto \exp[(T^*)^{-p}] \tag{11}$$

approximately holds if choosing $p = 2$. This leads to the classification of a fragile glass former (since $p > 1$) and the relaxation behavior is called Super-Arrhenius-like. The relaxation of the $MT^*_\text{ref}$-modeled systems is faster (which we might expect due to stronger diffusive motion) but also satisfies a Super-Arrhenius law, at least for the hot temperature regime and most parts of the supercooled regime. As expected, crystallization phenomena in the $MT^*_\text{ref}$-models, at quite low temperatures close to the glass transition temperature, are significantly represented in the failure of the Super-Arrhenius law at large simulation times within the framework of the so-called long sampling scheme (orange and purple solid lines in Fig. 10). Even at shorter times (orange and purple dashed lines), which we refer to as short sampling scheme, not a straight line is observed at low temperatures or large $(1/T^*)^2$ (i.e. Eq. (11).
Figure 10: (a) Alpha relaxation time $\tau_\alpha$ defined through Eq. (10) as a function of the inverse squared temperature (in dimensionless form $T^* = k_B T/\epsilon$) for the bidisperse Kob-Andersen system (solid black), the effective monodisperse system (black-dashed; corresponding fit added as a red curve) and the M0.45 and M0.9 modeled systems (purple; orange). Curves corresponding to a short sampling scheme are added for the latter models (colored-dashed). (b) Log-log plot of $\tau_\alpha$ as a function of $T^* - T_c^*$ for the bidisperse Kob-Andersen system (with $T_c^* = 0.435$), the M0.45 modeled system (with $T_c^* = 0.27$), M0.9 modeled system (with $T_c^* = 0.41$). The tilted dashed lines represent orientation lines being multiples of 10 of $\tau_\alpha^* = (T^* - T_c^*)^{-2.4}$. The horizontal dashed line marks the $\tau_\alpha$ value of the bidisperse KA system at the melting point.
does not hold). From the concave curved shape of the dashed curves, we can interpret that at low temperatures the system still fulfills the Super-Arrhenius law since each of the last tracked point before the glass temperature lies again on the straight curve of the respective $M_{T_{ref}}^*$ model. This can be explained by the fact that establishing a crystalline phase takes far more time at low temperatures such that the system remains in the $\alpha$-relaxation regime and shows only minor signs of an elongated relaxation which results from a crystallization in its initial process. Concerning the effective monodisperse system, we do not observe a Super-Arrhenius law; instead, we could fit a curve (red line) with $p = 0.2$. Accordingly, we yield a Sub-Arrhenius behavior for this non-glass forming model system. In these kinds of systems glassy energy barriers decrease towards lower temperatures since also the pair potential decreases when lowering temperature as shown in Fig. 5(b).

Kob and Anderson\textsuperscript{17} used the power law fit $\tau_\alpha^* \propto (T^* - T_c^*)^{-\gamma}$ to calculate a critical temperature $T_c^*$ (which resembles the glass transition temperature) and a critical exponent $\gamma$ to describe the transition from the ergodic regime with $T > T_c$ towards a nonergodic one with $T \leq T_c$. In this work we are interested in whether such a power law also holds for our $M_{T_{ref}}^*$ models close to their respective glass transition temperature and provide graphical results in Fig. 10(b). Firstly with respect to the bidisperse system, we observe the same power law behavior with $\gamma \approx 2.4$ close to $T_c^{\text{bidisperse}} = 0.435$ (as indicated by a straight line in Fig. 10(b)) as presented in the work of Kim and Saito\textsuperscript{20} resulting from an $F_s$ defined only among the A-particles (original work:\textsuperscript{10,17} $\gamma \approx 2.5(2.6)$ for A(B) particles). In the effective monodisperse system, however, no critical temperature can be extrapolated which is why it is not displayed. Fortunately, the $M_{T_{ref}}^*$ models each possess a glass transition temperature and yield about the same power law exponent as the bidisperse system. The relaxation times of these models are also faster close to their glass transition temperature than the relaxation times of the bidisperse system at the same temperature distance.

We next investigate how the diffusion coefficient $D$ depends on the alpha relaxation time $\tau_\alpha$ and temperature $T$. From Eq. (11) and also from previous paragraph, we know that
$\tau_a = \tau_a(T)$ approximately holds for all considered models revealing two different values for the coefficient $p$. In order to interpret our upcoming results, we first like to motivate our expectations. If, for example, we consider one large particle that moves slowly with respect to other–ideally smaller–ones, who form a viscous medium, the diffusion coefficient $D$ in terms of the other quantities $\tau_a$ and $T$ can be expressed via the Stokes-Einstein (SE) relation, that is

$$D = \frac{k_B T}{6\pi a \eta}$$

with $a$ and $\eta$ being the Stokes radius (the radius of the considered particle) and the shear viscosity, respectively. Literature devoted to the violation of the SE relation when supercooling a liquid can be found in Refs. 39–45. With respect to $\eta$, Debye provided an explicit expression in terms of $T$ and $\tau_a$, that is

$$\eta = \eta(T, \tau_a) = \frac{\tau_a k_B T}{4\pi a^3}.$$  

(13)

This expression simplifies Eq. (12) towards $D \propto \tau_a^{-1}$ under the assumption of a fixed (i.e. temperature invariant) Stokes radius $a$. However, this inverse relationship breaks down when supercooling the fluid.47,48 Other works investigating glassy systems,49–52 used a similar but fractional version of this expression, that is $D \propto (\tau_a)^{-\xi}$, to cover the dynamics in the highly supercooled regime. This coefficient $\xi$ is normally found to be slightly smaller than 1, whereas the ideal condition $\xi = 1$ would be consistent with Eqs. (12) and (13). In the following, we test such a power law expression in Fig. 11(a) by using a log-log plot for our modeled systems. We find that the slope, which is equal to $-\xi$, seems to differ from the ideal value of $-1$ for almost all of our models (a slope of -1 is parallel to the slim dashed lines), especially in the limit of a low $\tau_a$ (see inset in Fig 11(a)) as well as high $\tau_a$. These limits make the curves appear in a slightly convex shape. The effective monodisperse system reveals with $\xi = 1.05$ an almost ideal SE behavior although its value is slightly higher than 1 and the
Figure 11: (a) Diffusion coefficient $D$ versus alpha relaxation time $\tau_\alpha$. (b) Product of diffusion coefficient $D$ and alpha relaxation time $\tau_\alpha$ versus temperature $T$. Additional simulations for the bidisperse system for $1 < T^* \leq 1.5$ have been performed.
glass phase cannot be reached. With respect to the bidisperse system, we can detect at large \( \tau_\alpha \) (i.e. \( \tau_\alpha^* \gtrsim 10 \)) or equivalently low \( T \) (i.e. \( T^* \lesssim 0.6 \)) that with \( \xi = 0.78 \) the ideal value is significantly underrated. This is expected since we already know from the A- as well as the B-particles’ diffusive motion that a break-down of the SE relation towards a fractional one with \( \xi \approx 0.8 \) for A-particles, and \( \xi \approx 0.65 \) for B-particles is happening in the deeply supercooled regime (Ref. 16) determined by combining results from \( D \propto (T - T_c)^\gamma_1 \) and \( \tau_\alpha \propto (T - T_c)^{-\gamma_2} \). In order to have a broader picture of the entire \( D-\tau_\alpha-T \) relationship, we present in Fig. 11(b) a “\( D\tau_\alpha^* \)” versus “\( T \)” plot, in which a strong \( T \) or \( \tau_\alpha \) dependence of \( D \times \tau_\alpha \) at \( T^* \lesssim 0.6 \) for the bidisperse system (black-solid) indicates this breakdown. Nonetheless, consistency for that model with the SE relation is found at an intermediate temperature regime (0.7 \( \lesssim T^* \lesssim 1 \)) before reaching the melting point beyond which \( \xi(T^* \gtrsim 1) = 1.19 \) holds and a linear growth of \( D \times \tau_\alpha \) with \( T \), i.e. \( D \tau_\alpha \propto T \), is found (inset of Fig. 11(b)). This means that also at high temperatures the diffusion is stronger than expected with respect to the structural relaxation which is caused by a slight growth in the Stokes radius. Such linear relationship is obtained from Eq. (12) if the shear viscosity \( \eta \) is related to \( \tau_\alpha \) such that

\[
\eta = \eta(\tau_\alpha) \propto \tau_\alpha \tag{14}
\]

and was already observed in supercooled fluids, even in fractional form. This dynamic characteristics is also found in our M0.45 and M0.9 models. Even more, the temperature at which we denote in the respective \( M_{T_{ref}} \) model the change in the dynamics, approximately coincides with the temperature at which we denote the same relaxation time as in the bidisperse system (see Fig. 10 showing these temperatures whose \( \tau_\alpha \) values lie on the same horizontal line).

Investigations involving these two versions of the SE relation can be found in Refs. 57,58 whereas generalized versions of this relation are investigated in Refs. 56,59. The M0.45 and M0.9 models’ \( \xi \) coefficients possess in their high temperature regimes with \( \xi_{M0.45}(T^* > 27) \)
0.45) = 1.35 and $\xi_{M0.9}(T^* > 0.7) = 1.22$, respectively, a higher as well as similar value compared to the reference, but still and also in accordance to the bidisperse system, the relation $\xi < 1$ for temperatures $T \rightarrow T_c$ holds ($\xi_{M0.45}(T^* < 0.35) = 0.79$, $\xi_{M0.9}(T^* < 0.6) = 0.8$). Since corresponding potentials are fixed, the weakening with respect to the thermal energy occurs in a reciprocal way to temperature. As a consequence, the M0.45 and the M0.9 models’ change in pair potential magnitude is far more sensitive with respect to temperature than the weakening of the intrinsic temperature dependent effective pair potential in units of the thermal energy as depicted in Fig 12. We conclude that it must be this growing influence of the undulating pair potential at lower temperatures that leads to increased Stokes radii and thus to the existence of a glass transition.

![Figure 12: Pair potentials for the M0.9 model (solid lines) and the effective monodisperse model (dashed lines) in units of $k_B T$ at the (dimensionless) temperatures $T^* = 0.3, 0.6, 0.9$.](image)

Figure 12: Pair potentials for the M0.9 model (solid lines) and the effective monodisperse model (dashed lines) in units of $k_B T$ at the (dimensionless) temperatures $T^* = 0.3, 0.6, 0.9$. Both models coincide at $T^* = 0.9$ by definition.

More insights into the dynamics can be obtained by analyzing the particle’s activity via excitation events. These events are characterized by persistence and exchange time that are defined as follows: the persistence time $\tau_p$ for a particle $i$ is the minimal waiting time $t_1$ to undergo its first excitation such that $\|r_i(t_1) - r_i(0)\| \geq d$ holds, whereas the exchange time $\tau_x$ represents the time between subsequent excitation events, i.e. $\|r_i(t_2 + t_1) - r_i(t_1)\| \geq d$, $\|r_i(t_3 + t_2 + t_1) - r_i(t_1 + t_2)\| \geq d$, etc. The parameter $d$ represents the critical displacement for which we chose $d = \sigma$ for all considered models.

By capturing a lot of $\tau_p$ and $\tau_x$ values, corresponding distributions can be created. A
full alignment between the two distributions means that the probability distribution of the
time length, an arbitrarily chosen particle at an arbitrarily chosen simulation time needs to
undergo a full excitation event, is equal to those of a particle which just went through such
event. This seems plausible at high temperatures at which there is a very short structural
relaxation time $\tau_\alpha$ leading quickly to equal mobility characteristics throughout the system.
If, however, $\tau_\alpha$ becomes more dominant in relation to $\langle \tau_p \rangle := \mathbb{E}(P(\tau_p))$ local structures in
configurational and momentum space that have favored a prior excitation event might still
exist and more easily tend to trigger another event. As a result, the system can exhibit
dynamic heterogeneity which is a phenomenon describing the spatial partition of the sys-
tem in dynamically active and inactive regions, i.e. regions in which the average particles’
displacement is significantly altered from those of the system’s average. In Fig. 13 the dis-
tributions $P(\tau_p)$ and $P(\tau_x)$ and their first moment ratios $\langle \tau_p \rangle / \langle \tau_x \rangle$ are depicted covering
in Subfigs. (a,b) the bidisperse system, (c,d) the effective monodisperse system as well as
in (e,f) the system modeled via the M0.9 model for the short sampling scheme. With re-
spect to the bidisperse system (Fig. 13(a,b)), alignment between $P(\tau_p)$ and $P(\tau_x)$ is found
at high temperatures, e.g., $T^* \gtrsim 0.7$. However, even at high temperatures above the glass
transition temperature, e.g., $T^* \approx 0.6$, alignment quickly disappears upon cooling leading
to persistence times far above excitation times. That means, when cooling the system, a
particle needs a lot of time to get activated, whereas once being activated, it tends more and
more to remain in that state. As a result, the first moment ratio $\langle \tau_p \rangle / \langle \tau_x \rangle$ significantly
increases with respect to the ideal value of 1 upon cooling as depicted in Subfig. 13(b). In the
effective monodisperse system (Fig. 13(c,d)), the distribution alignment holds even at lower
temperatures and thus preventing the ratio $\langle \tau_p \rangle / \langle \tau_x \rangle$ to diverge. The M0.9-modeled system
(Fig. 13(e,f)), on contrary, possesses a similar characteristics as those of the bidisperse sys-
tem as long as we constrain the analysis to the short sampling scheme at which we have not
a progressed crystallization process. In Fig. 13(f) we provide also the curve resulting from
the long sampling scheme. From that graph, we can also see that from a temperature of
Figure 13: Distributions of the persistence and exchange time and their first moment ratios $\langle \tau_p \rangle / \langle \tau_x \rangle$ for (a,b) the bidisperse system, (c,d) the effective monodisperse system as well as (e,f) the system modeled via the M0.9 model at various temperatures (in dimensionless form: $T^* = k_B T/\epsilon$). For visual simplicity, the distributions were rescaled as a function of temperature. Subfigs. (a)-(d) display results stemming from the long sampling scheme. Subfigs. (e,f) display results from the short sampling scheme and (f) additionally from the long sampling scheme.
\( T^* \approx 0.6 \) onward, we have full alignment between both equilibrium sampling schemes.

In order to further quantify dynamic heterogeneity, we focus in Fig. 14 on the rescaled variance of the \( \hat{F}_s \) function defined in Eq. (9), which is also called four-point dynamic susceptibility function, and defined through

\[
\chi_4(k,t) = N \cdot \left( \left\langle \hat{F}_s(k,t)^2 \right\rangle - \left\langle \hat{F}_s(k,t) \right\rangle^2 \right).
\]

(15)

Its maximum value is by definition located where the the dynamic fluctuation of \( \hat{F}_s \) is maximized and the positions of the maxima follow the alpha relaxation time, which we have displayed in Fig. 10. We observe in the bidisperse system (Fig. 14(a)), that the position of the maximum of \( \chi_4 \) is growing by lowering the temperature. This is expected since we already know that a similar result was found in a former investigation of the dynamics in the bidisperse KA system. However, we hereby focus on all particles rather than species A alone.

In the effective monodisperse system (Fig. 14(b)), we also observe a growth of the maximum of \( \chi_4 \) but the maxima of neighboring curves only slightly differ, meaning that dynamic heterogeneity is not a dominant artifact being observed in this low-fragility \( (p = 0.2) \) fluid while cooling. This is consistent with a prior investigation in which it was observed that more fragility leads to more dynamic heterogeneity. In the remaining Subfig. (c), we present \( \chi_4 \)-results for the M0.9 model for the long, and additionally, for the short sampling scheme. Clearly and in a similar fashion to the bidisperse system, we can detect a significant growth of \( \chi_4 \)'s maximum corresponding to strongly increasing dynamic heterogeneity upon cooling when considering the short sampling scheme. As expected, we detect a strong alignment between curves from both sampling schemes at temperatures above the glass transition temperature at which no crystallization occurred within the framework our investigation (i.e. \( T^* > 0.5 \)).

As a results of our study, we can obtain an effective monodisperse system out of a
Figure 14: Dynamic susceptibility $\chi_4$ for a broad range of temperatures (in dimensionless form: $T^* = k_B T/\epsilon$) for (a) the original Kob-Andersen system, (b) the effective monodisperse system and (c) the M0.9-modeled system. Subfig. (c) contains also results for the short sampling scheme (dashed curves).
bidisperse, or generally multidisperse system, that has similar dynamic features. In our case, it is possible to yield an effective model with fast relaxation that has also a similar glass transition temperature. However, the implementation of the desired dynamical features led to minor trade-offs in the structural similarity as displayed in Fig. 7.

4 Conclusion

In this article, we presented a coarse-graining method aiming to reduce the number of particle species or labels. Hereby the well-known bidisperse Kob-Andersen mixture was transformed into a monodisperse system at the same particle density and coarse-grained by forcing the all-particle radial distribution function being invariant in both models. The corresponding effective pair potential, which is temperature and density dependent, was developed using the iterative Boltzmann inversion scheme.

We found that in the effective system the diffusion coefficient has quite high values at low temperatures in comparison to the bidisperse model (see Fig. 8). These high values lead to an absence of the glass transition in the effective system. It results from a reduction in potential strength among particle pairs when lowering the temperature (Fig. 5(b)) and makes it easier for particles to re-position and diffuse on a large time scale. In order to reintroduce the ability to appear in a glassy state, we considered also two models having no intrinsic temperature dependence. These models each correspond to the temperature dependent effective monodisperse model at a specific dimensionless temperature $T_{\text{ref}}^*$ and were denoted with M0.45 and M0.9 (corresponding to $T_{\text{ref}}^* = 0.45, 0.9$). By using this type of approach it was possible to obtain a glass transition temperature if we start from the equilibrium configuration at $T_{\text{ref}}^*$ and instantly use it at the desired temperature for a short time, i.e. short enough that the system cannot crystallize but still long enough to well reach the plateau of the alpha relaxation regime (see Fig. 9). We further identified that the glass transition temperature can be changed by taking the right choice for $T_{\text{ref}}^*$, which was found
being around 0.9.

A major result of our investigation is that we can effectively reproduce a bidisperse fluid with a monodisperse one if we make minor concessions towards marginal structural changes which, fortunately, retain the isotropic property.

Our findings suggest a possible use for designing or manufacturing a less or even one-component type of colloidal model fluid out of complex fluids such as paints or lubricants. Resulting one-component fluids might be even capable to remain in the liquid state for a broad parameter range, i.e. also in regions where an unwanted eutectic phase separation would occur in the corresponding multidisperse system. However, designing such effective particles might be difficult or even impossible for certain systems since the effective interactions might be too complex for manufacturing the corresponding colloids or nanoparticles. Another useful system of application is given through ionic liquids which are usually described through bidisperse fluids possessing glassy dynamics. Each ionic particle consists of one or more charged coarse-grained sites. Computer simulations in such a system normally have the drawback that evolving the equations of motion is quite involved due to long-range electrostatic interactions. By considering a fully mixed neutral ionic liquid, we would effectively cover the screening effect with our method. The screening might shorten the range of the effective potential compared to the Coulomb potential and thus speeding-up a computer simulation.

Finally, we would like to point out that the investigation so far considers only the interparticle distance as a variable to describe a pair configuration. Perhaps, one might extend such an investigation towards more complex pair configuration descriptions.

**Conflict of interest**

There are no conflicts to declare.
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