A Coarse-Graining Procedure for Mapping Atomistic Models

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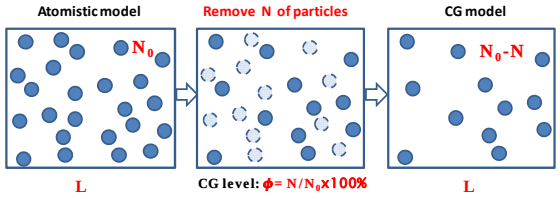
Motivation

Continuum mechanics: *efficient but inadequate at nano scales.*
Molecular dynamics: *accurate but computationally expensive.*
Goal: *to develop effective hybrid computational methods for nano-scale flows.*
Key issue: *to determine whether an intermediate mesoscale description is required.*

Model

Atomistic model: $N_0 = 1000$, ρ(number density) $= 0.8$, $r_{cut} = 2.5\sigma$ and Lennard-Jones (LJ) potential.

$$U^{LJ}(r) = 4\varepsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r}\right)^{12} - \left(\frac{\sigma_{ij}}{r}\right)^{6}\right]$$



Coarse-grained (CG) model should preserve the thermodynamic and dynamic properties of the detailed atomistic system.

Method

Iterative Boltzmann inversion: *construct an effective pairwise potential for the CG model that reproduces the radial distribution function, g(r), of the detailed atomistic system.*[1]

$$U_0 = -k_B T \ln g_{target}(r)$$

$$U_{i+1}(r) = U_i(r) + \xi k_B T \ln\left[\frac{g_i(r)}{g_{target}(r)}\right]$$

Pressure matching: *put pressure constraint on the effective potential through an optimization procedure.*

➢ Objective function

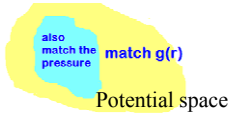
$$f[U] = \frac{1}{2}\int_0^{cutoff}\left(\ln\frac{g_i(r,U)}{g_{target}(r)}\right)^2 dr + \frac{1}{2}\alpha\left(\frac{P_{CG}}{P_{AA}} - 1\right)^2$$

(Penalty parameter; structure; Pressure (penalty function))

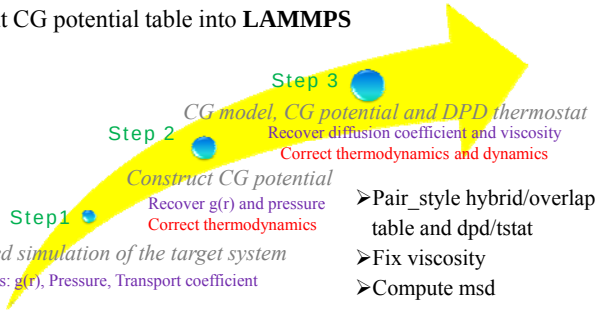
➢ Steepest decent algorithm

$$u_j^{k+1} = u_j^k - \frac{\partial f^k}{\partial u_j^k}\chi$$

(Step size)



➢Input CG potential table into **LAMMPS**



➢Pair_style hybrid/overlap table and dpd/tstat
➢Fix viscosity
➢Compute msd

Dissipative Particle Dynamics thermostat (DPD)[2]

➢Galilean-invariant thermostat
➢ the dissipative force term mimics the friction between particles and dissipates energy.

$$\dot{\vec{p}}_i = \vec{F}_i^C + \vec{F}_i^D + \vec{F}_i^R$$

$$\vec{F}_i^D = \sum_{j\neq i}\vec{F}_{ij}^D,\quad \vec{F}_{ij}^D = -\gamma w^D(r_{ij})(\hat{r}_{ij}\cdot\hat{v}_{ij})\hat{r}_{ij}$$

$$\vec{F}_i^R = \sum_{j\neq i}\vec{F}_{ij}^R,\quad \vec{F}_{ij}^R = -\sigma w^R(r_{ij})\Theta_{ij}\hat{r}_{ij}$$

Viscosity: Reverse Nonequilibrium Molecular Dynamics (RNEMD) [3]

$$j_z(p_x) = -\eta\frac{\partial v_x}{\partial z},\quad j(p_x) = \frac{P_x}{2tA}$$

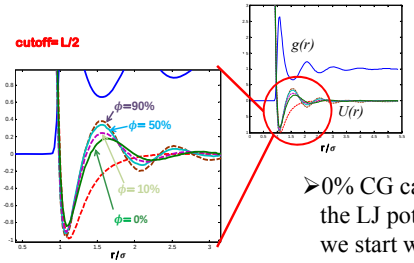
➢linear response theory

Diffusion: Mean Square Displacement (MSD)

$$D = \frac{1}{6t}\sum_{\alpha=1}^{3}\left\langle\left[r_{\alpha i}(t) - r_{\alpha i}(0)\right]^2\right\rangle$$

Results and Discussion

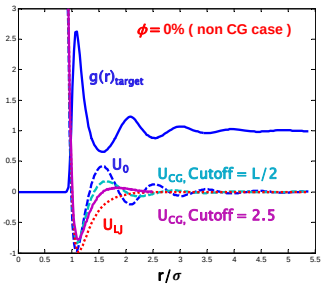
Effective potentials for various CG levels



➢Potential of mean force is a good initial guess for high CG level case.

➢0% CG case doesn't reproduce the LJ potential($r_{cut} = 2.5$) since we start with a longer-ranged interaction($r_{cut} = L/2 \approx 5.4$).

Convergence test[1]

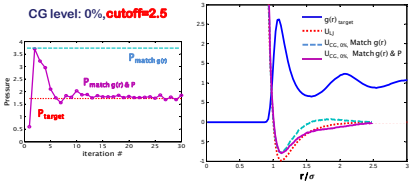


➢The potential is closer to the LJ potential when r_{cut} is reduced to 2.5.

➢the structure of a dense LJ ($\rho \gtrsim 0.65$) system is mainly determined by the repulsive part of the potential.[4]

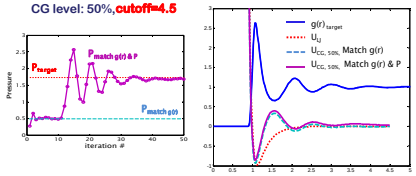
After matching the pressure

➢The attractive part of the potential is recovered. Now the potentials are more similar qualitatively.



➢Pressure matching acts mainly on the long-range potential.

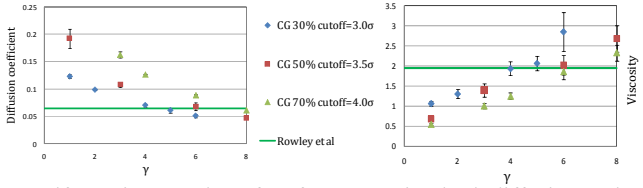
➢Pressure is very sensitive to the potential, unlike g(r).



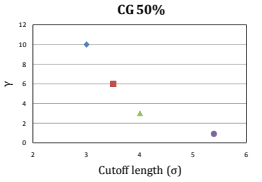
➢Long-ranged interactions are important in high CG level case. The cutoff needs to be extended to recover g(r) and pressure.

➢The cutoff should be optimized based on the computational efficiency.

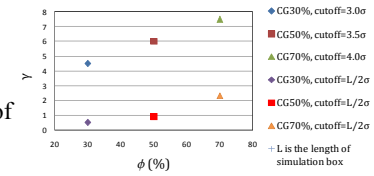
Match the dynamical properties



➢A self-consistent value of γ for recovering both diffusion and viscosity is obtained for each CG level with the optimized cutoff length.



➢The friction coefficient, γ, should increase as the length of interaction decreases.



➢γ increases as the degree of CG (ϕ) increases.

References

[1] Reith, D. et al., *J. Comput. Chem.* 24, no. 13 (April 2003): 1624-1636.
[2] Groot, R. D. and Warren, P. B., *J. Chem. Phys.* 107, no. 11 (June 1997): 4423-4435.
[3] Muller-Plathe, F., *Phys. Rev. E.* 59, no. 5 (May 1999): 4894-4898.
[4] Weeks, J. D. et al., *J. Chem. Phys.* 54, no. 12 (June 1971): 5237-5247.