

## Supporting documentation for

### *Comparison of equilibrium techniques for the viscosity calculation from DPD simulations*

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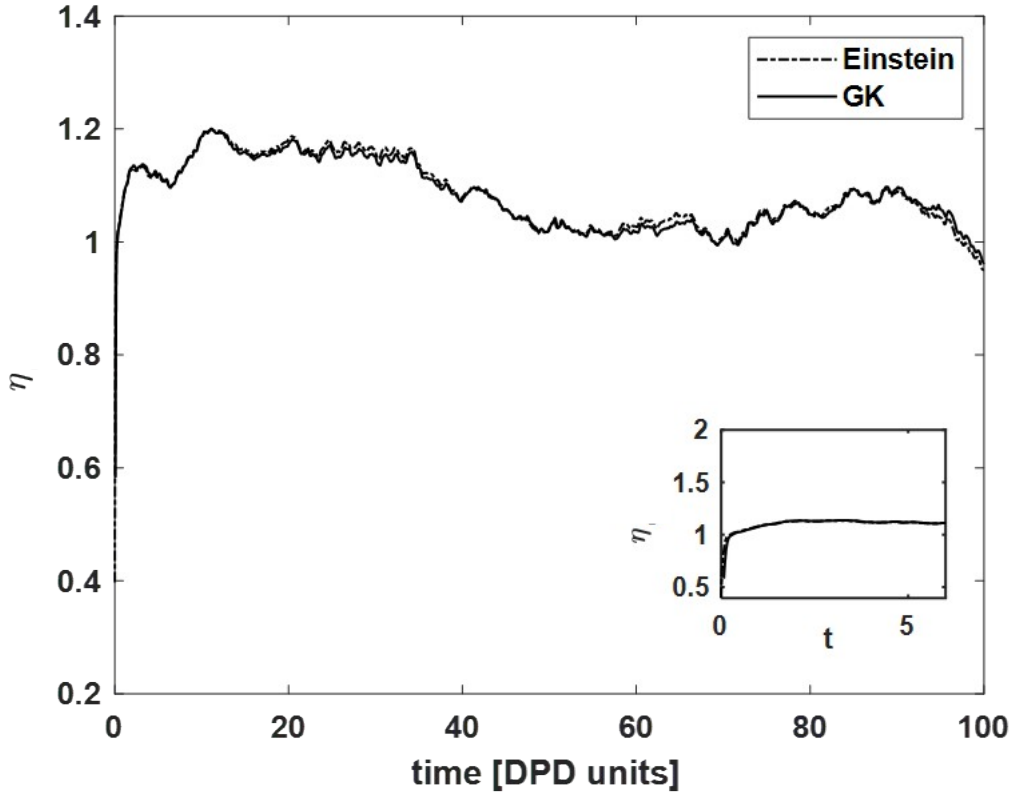
## 1. Numerical validation of revised Einstein formula in DPD water system

The derived Einstein formula (eq. 2) was validated numerically and compared with the revised GK formula suggested by Jung and Schmid [1] (eq. 1) in water DPD system, *i.e.* the conservative parameter was  $a_{ij} = 25$  and  $\gamma = 4.5$ . The results are shown in Figure S 1 and the error between the two methods is found to be at 0.8%. This error is obtained after a single run on a system of 192000 DPD water particles and a box size of  $L^3 = 40^3$ . The timestep of integration is  $dt = 0.02$ .

$$\eta_{\alpha\beta}^{GK} = \eta_{\infty}^{GK} + \frac{V}{k_B T} \int_0^{\infty} \langle (P_{\alpha\beta}^P(t_0) - P_{\alpha\beta}^D(t_0))(P_{\alpha\beta}^P(t_0 + t) + P_{\alpha\beta}^D(t_0 + t)) \rangle_{t_0} dt \quad (1)$$

where  $\eta_{\infty}^{GK} = \frac{V}{k_B T} \frac{\Delta t}{2} \langle P_{\alpha\beta}^{R2} \rangle$ .

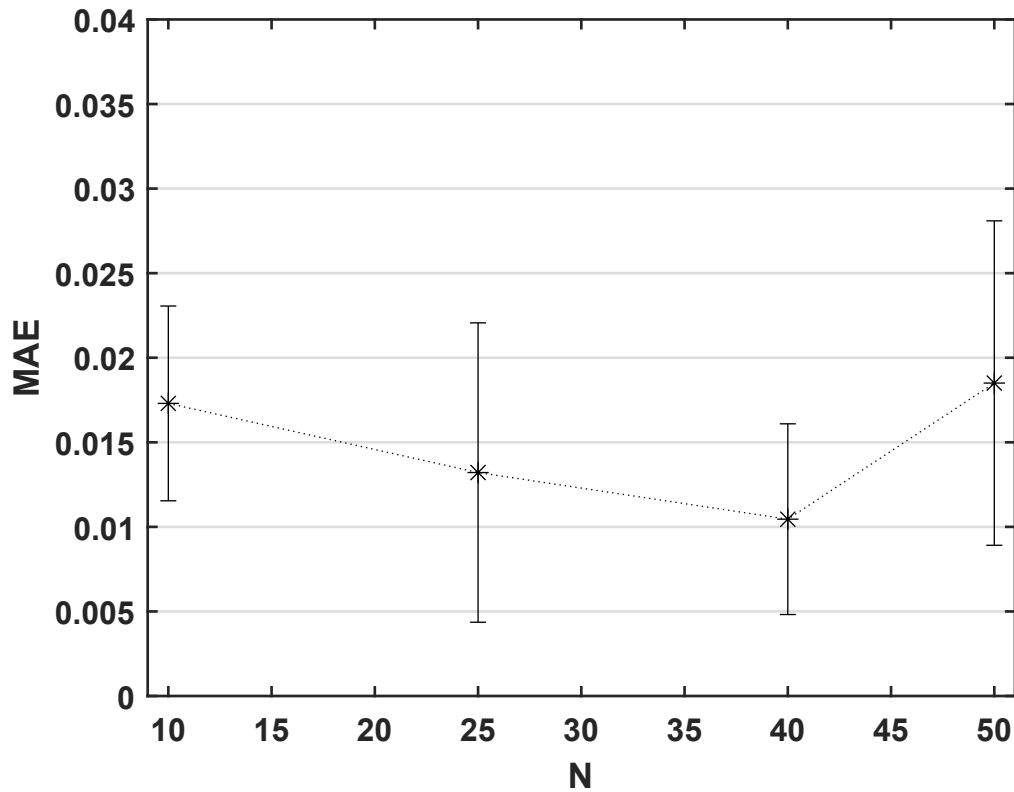
$$\eta_{\alpha\beta}^E = \lim_{t \rightarrow \infty} \frac{V}{2k_B T} \frac{d}{dt} \langle \eta_{\alpha\beta}^{PP} - \eta_{\alpha\beta}^{DD} \rangle_{t_0} + \frac{V}{k_B T} \lim_{t \rightarrow 0} \frac{1}{t} \langle \eta_{\alpha\beta}^{RR} \rangle_{t_0} \quad (2)$$



**Figure S 1: Comparison of zero-shear viscosity from revised GK and Einstein methods. The zero-shear viscosity in the time interval  $[0, 5.5]$  is illustrated in the inset figure.**

The DPD water system, *i.e.*  $a_{ij} = 25, \gamma = 4.5$  is expected to equilibrate quickly due to the single type particles that are not bonded with each other. Thus, the viscosity cut off point can be taken after a relatively short period of time. In our case we see that the viscosity equilibrates

around unit if  $t < 5$  while for  $t \sim 0.8$  it takes values close to 1.1 which is in agreement with previous works [2,3] and with the non-equilibrium Lees- Edwards method that we applied.



**Figure S 2: Mean Absolute Error (MAE) between the average viscosities calculated using the GK and Einstein relations as a function of the polymer chain lengths. The average for each polymer length,  $N$ , is taken over every polymer concentration and  $a_{sp}, \gamma_{sp}$  values.**

## 2. Mean square end-to-end distance and characteristic ratio

The end-to-end distance in polymer melts follows the same trend for each conservative and dissipative parameter as the average radius of gyration (Figure 1 in main article).

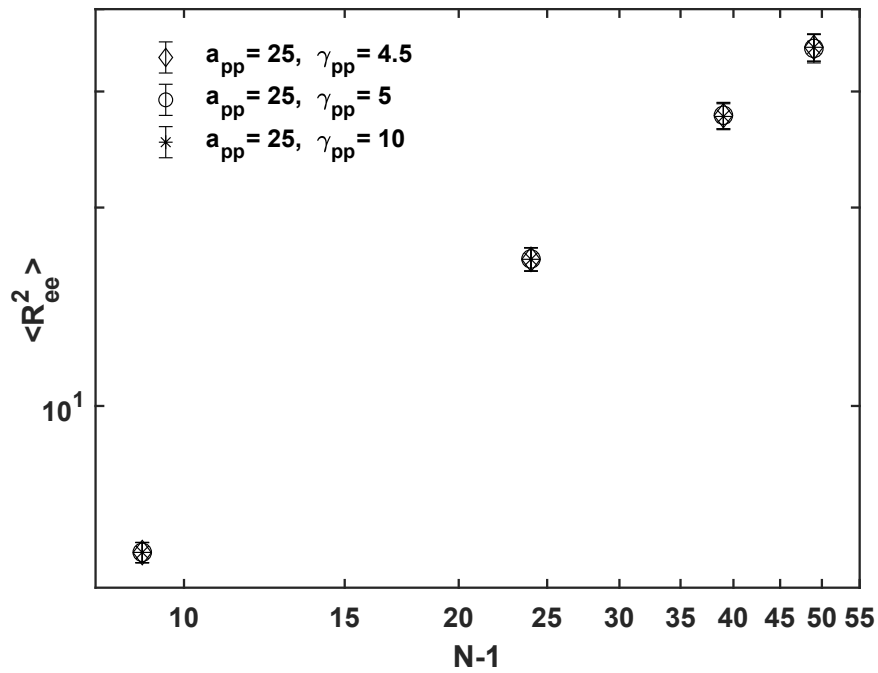


Figure S 3: Mean square end-to-end distance as a function of polymer length and interaction parameters. The error bars correspond to the standard deviation of the mean value over the time frames.

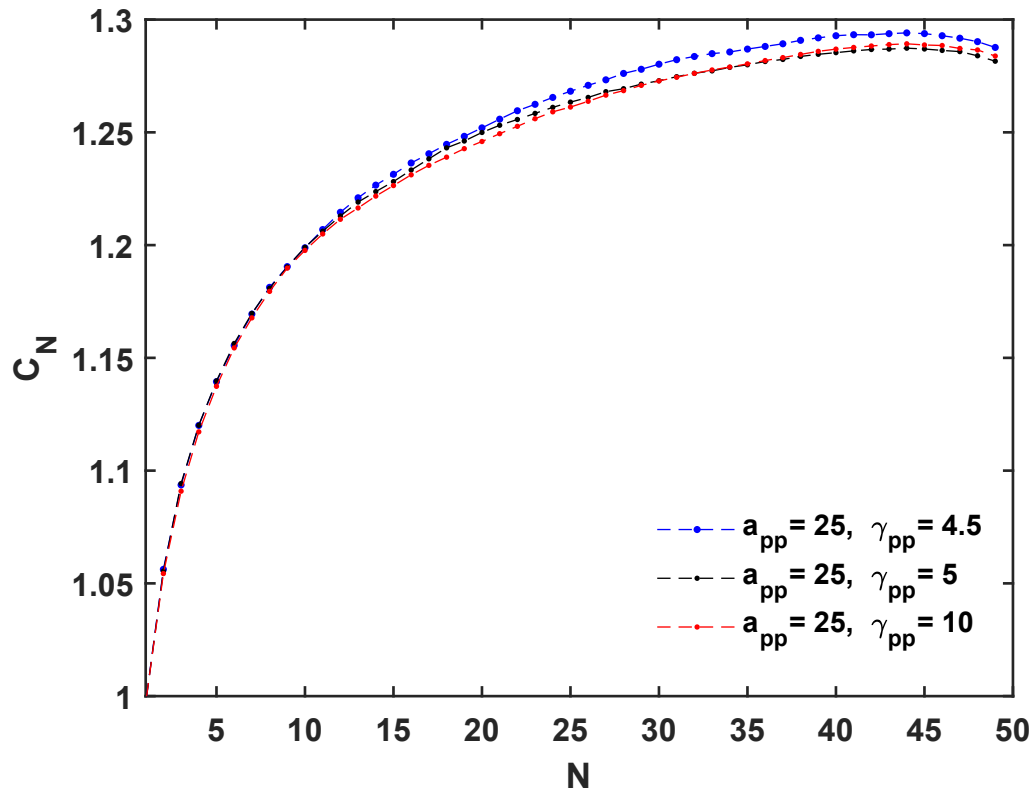


Figure S 4: Characteristic ratio,  $C_N = \frac{R_{ee}^2}{(N-1)l_b^2}$ , where  $l_b^2$  is the mean squared bond length, calculated for polymer melts as a function of polymer length and interaction parameters.



### 3. Random selection of end-to-end vector autocorrelation figures

The autocorrelation function of the end-to-end vector shown in Figure S4 has the same trend for each conservative and dissipative parameter studied in this work. The vector decorrelates faster the shorter the polymer chains are. As expected, it is observed that the slowest decorrelation of  $\vec{e}\vec{e}$  is shown by the longest polymer, i.e.,  $N = 50$ .

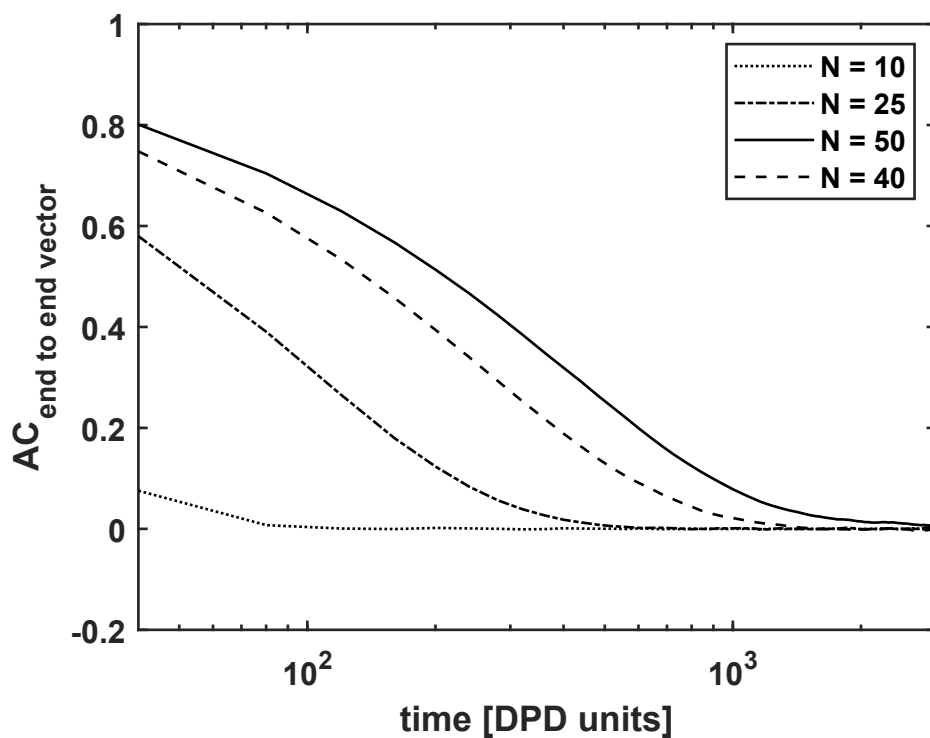


Figure S 5: Autocorrelation function of end-to-end vector at  $a_{sp} = 15, \gamma_{sp} = 4.5$  and 30% polymer concentration, i.e.  $c_p = 0.3$

#### 4. Parabolic fitting on Schmidt number

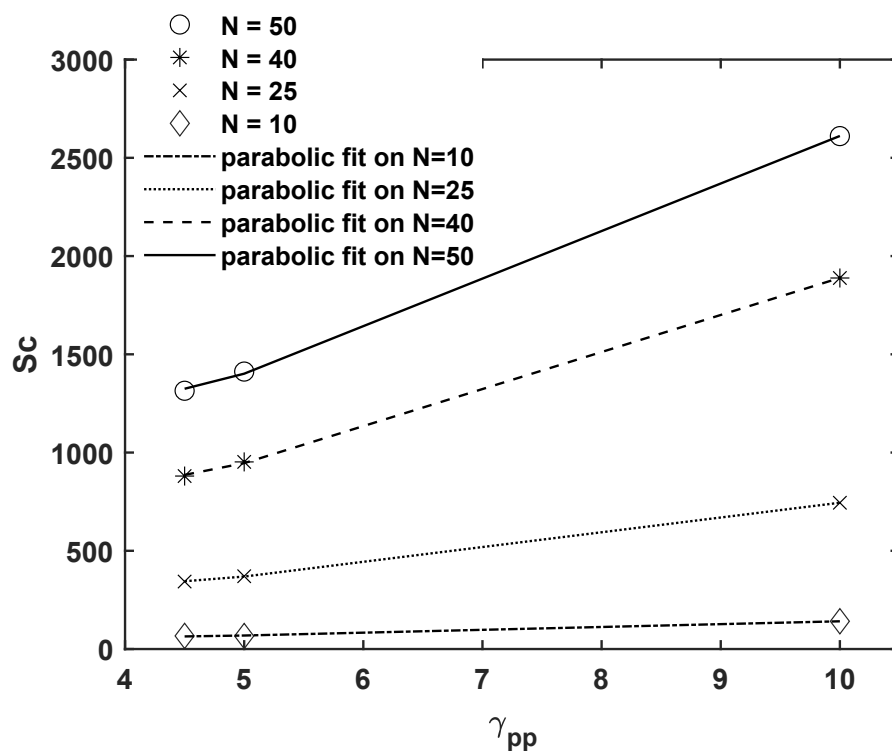
The Schmidt numbers are plotted against the friction interaction parameter  $\gamma$  and then fitted using the next equation:

$$A + \frac{(2\pi\gamma_{ij}\rho r_c^4)^2}{Bm^2k_B T}$$

where  $A, B$  are fitting coefficients. The fitting coefficients that were obtained are given in table S1.

**Table S 1: Fitting coefficients from the parabolic fitting on Schmidt numbers**

N	A	B
10	44.67	368.10
25	243.63	70.92
40	631.85	28.27
50	998.05	22.02



**Figure S 6: Parabolic fitting on the Schmidt number as a function of friction parameter. The Schmidt numbers shown in this figure correspond to the polymer melt case (Fig. 9  $c_p = 1$  in the main manuscript).**

## 5. References

- [1] G. Jung and F. Schmid, *Computing Bulk and Shear Viscosities from Simulations of Fluids with Dissipative and Stochastic Interactions*, J. Chem. Phys. **144**, 204104 (2016).
- [2] A. Chaudhri and J. R. Lukes, *Velocity and Stress Autocorrelation Decay in Isothermal Dissipative Particle Dynamics*, Phys. Rev. E - Stat. Nonlinear, Soft Matter Phys. **81**, 1 (2010).
- [3] J. A. Backer, C. P. Lowe, H. C. J. Hoefsloot, and P. D. Iedema, *Poiseuille Flow to Measure the Viscosity of Particle Model Fluids*, J. Chem. Phys. **122**, 154503 (2005).