

## 1 First Approximation

First term:

$$\frac{\partial \Psi^{(0)}}{\partial \rho_b} \frac{\partial \rho_b}{\partial t} = -\Psi^{(0)} \nabla_{\mathbf{r}} \cdot \mathbf{c}_0 - \frac{\Psi^{(0)} \mathbf{c}_0}{\rho_b} \cdot \nabla_{\mathbf{r}} \rho_b \quad (\text{S1})$$

Second term:

$$\begin{aligned} \frac{\partial \Psi^{(0)}}{\partial \mathbf{c}_0} \frac{\partial \mathbf{c}_0}{\partial t} &= \Psi^{(0)} \frac{m}{k_b T} (\mathbf{c} - \mathbf{c}_0) \cdot \left( -\frac{k_b T}{m} \nabla_{\mathbf{r}} T - \mathbf{c}_0 \nabla_{\mathbf{r}} \cdot \mathbf{c}_0 - \frac{k_b T}{m \rho_b} \nabla_{\mathbf{r}} \rho_b + \sum_j \frac{\rho_j \mathbf{F}_j}{m \rho_b} \right) \quad (\text{S2}) \\ &= -\Psi^{(0)} \mathbf{C} \cdot \nabla_{\mathbf{r}} \ln T - \Psi^{(0)} \frac{m}{k_b T} \mathbf{C} \cdot (\mathbf{c}_0 \nabla_{\mathbf{r}} \cdot \mathbf{c}_0) - \frac{\Psi^{(0)}}{\rho_b} \mathbf{C} \cdot \nabla_{\mathbf{r}} \rho_b + \Psi^{(0)} \mathbf{C} \cdot \sum_j \frac{\rho_j \mathbf{F}_j}{\rho_b k_b T} \quad (\text{S3}) \end{aligned}$$

Third term:

$$\frac{\partial \Psi^{(0)}}{\partial T} \frac{\partial T}{\partial t} = \frac{3}{2} \Psi^{(0)} \mathbf{c}_0 \cdot \nabla_{\mathbf{r}} \ln T + \Psi^{(0)} \nabla_{\mathbf{r}} \cdot \mathbf{c}_0 - \Psi^{(0)} W^2 \mathbf{c}_0 \cdot \nabla_{\mathbf{r}} \ln T - \Psi^{(0)} \frac{2}{3} W^2 \nabla_{\mathbf{r}} \cdot \mathbf{c}_0 \quad (\text{S4})$$

We have used  $W^2 = \frac{m}{2k_b T} (\mathbf{c} - \mathbf{c}_0)^2$ . Eventually, the last term:

$$\mathbf{c} \cdot \nabla_{\mathbf{r}} \Psi^{(0)} = \frac{\mathbf{c} \Psi^{(0)}}{\rho_b} \cdot \nabla_{\mathbf{r}} \rho_b - \frac{3}{2} \Psi^{(0)} \mathbf{c} \cdot \nabla_{\mathbf{r}} \ln T + \Psi^{(0)} W^2 \mathbf{c} \cdot \nabla_{\mathbf{r}} \ln T + 2\Psi^{(0)} \sqrt{\frac{m}{2k_b T}} \mathbf{W} \cdot (\mathbf{c} \nabla_{\mathbf{r}} \cdot \mathbf{c}_0) \quad (\text{S5})$$

To reiterate, we have let  $\mathbf{C} = \mathbf{c} - \mathbf{c}_0$ , this leads to:

$$\Psi^{(0)} \left( \left[ -\frac{5}{2} \mathbf{C} \cdot \nabla_{\mathbf{r}} \ln T + W^2 \mathbf{C} \cdot \nabla_{\mathbf{r}} \ln T \right] + 2[\mathbf{W}\mathbf{W} : \nabla_{\mathbf{r}} \mathbf{c}_0 - \frac{1}{3} W^2 \mathbf{I} : \nabla_{\mathbf{r}} \mathbf{c}_0] + \mathbf{C} \cdot \sum_j \frac{\rho_j \mathbf{F}_j}{\rho_b k_b T} \right) = -\frac{\Psi^{(1)} - \Psi^{(0)}}{\tau} \quad (\text{S6})$$

$$\Psi^{(1)} = \Psi^{(0)} \left[ 1 - \tau \left[ \left( -\frac{5}{2} \mathbf{C} \cdot \nabla_{\mathbf{r}} \ln T + W^2 \mathbf{C} \cdot \nabla_{\mathbf{r}} \ln T \right) + 2(\mathbf{W}\mathbf{W} : \nabla_{\mathbf{r}} \mathbf{c}_0 - \frac{1}{3} W^2 \mathbf{I} : \nabla_{\mathbf{r}} \mathbf{c}_0) + \mathbf{C} \cdot \sum_j \frac{\rho_j \mathbf{F}_j}{\rho_b k_b T} \right] \right] \quad (\text{S7})$$

$\mathbf{I}$  is a  $3 \times 3$  identity matrix.  $\rho_j$  is the number density of the  $j$ th bead such that  $\rho_b = \sum_j \rho_j$  and  $\mathbf{F}_j$  is the intramolecular force acting upon the  $j$ th bead.

## 2 Evaluation of the Component $p_{xy}$

$C$  is the magnitude of the vector  $\mathbf{C}$ ,  $C_x$  and  $C_y$  are the  $x$  and  $y$  components of the vector  $\mathbf{C}$ . To be more explicit,

$$\mathbf{C} = \begin{bmatrix} C_x \\ C_y \\ C_z \end{bmatrix} \quad (\text{S8})$$

$$C = \sqrt{C_x^2 + C_y^2 + C_z^2} \quad (\text{S9})$$

In spherical coordinates, we have:

$$\mathbf{C} = \begin{bmatrix} C \sin \theta \cos \psi \\ C \sin \theta \sin \psi \\ C \cos \theta \end{bmatrix} \quad (\text{S10})$$

Then:

$$\int_{-\infty}^{\infty} C_x C_y \Psi^{(0)} d\mathbf{C} = \int_0^{2\pi} \sin \psi \cos \psi d\psi \int_0^{\pi} \sin^3 \theta d\theta \int_0^{\infty} C^4 \Psi^{(0)} dC \quad (\text{S11})$$

The integral  $\int_0^{2\pi} \sin \psi \cos \psi d\psi = 0$ , therefore:

$$\int_{-\infty}^{\infty} C_x C_y \Psi^{(0)} d\mathbf{C} = 0 \quad (\text{S12})$$

Now, consider the terms:

$$\int_{-\infty}^{\infty} C_x C_y \left(-\frac{5}{2}\mathbf{C} + W^2\mathbf{C}\right) \Psi^{(0)} d\mathbf{C} \cdot \nabla_{\mathbf{r}} \ln T \quad (\text{S13})$$

And:

$$\int_{-\infty}^{\infty} C_x C_y \mathbf{C} \Psi^{(0)} d\mathbf{C} \cdot \sum_j \frac{\rho_j \mathbf{F}_j}{\rho_b k_b T} \quad (\text{S14})$$

By considering terms in three different directions:

$$\left(W^2 - \frac{5}{2}\right) \frac{\partial \ln T}{\partial l} \int_0^{2\pi} \int_0^{\pi} \int_0^{\infty} C_x C_y C_l \Psi^{(0)} C^2 dC \sin \theta d\theta d\psi \quad (\text{S15})$$

$$\left(\sum_j \frac{\rho_j F_{j,l}}{\rho_b k_b T}\right) \int_0^{2\pi} \int_0^{\pi} \int_0^{\infty} C_x C_y C_l \Psi^{(0)} C^2 dC \sin \theta d\theta d\psi \quad (\text{S16})$$

where  $l$  can be  $x$ ,  $y$  or  $z$ .  $F_{j,l}$  is the intramolecular force acting upon  $j$ th bead in the  $l$  direction. Consider the integral over  $\psi$ , by the fact that  $C_x = C \sin \theta \cos \psi$ ,  $C_y = C \sin \theta \sin \psi$ , and  $C_z = C \cos \theta$ , we only have to confirm that the followings are zeros:

$$\int_0^{2\pi} \cos^2 \psi \sin \psi d\psi = -\frac{1}{3} \left[ \cos^3 \psi \right]_0^{2\pi} = 0 \quad (\text{for, } l = x) \quad (\text{S17})$$

$$\int_0^{2\pi} \cos \psi \sin^2 \psi d\psi = \frac{1}{3} \left[ \sin^3 \psi \right]_0^{2\pi} = 0 \quad (\text{for, } l = y) \quad (\text{S18})$$

$$\int_0^{2\pi} \cos \psi \sin \psi d\psi = -\frac{1}{2} \left[ \cos^2 \psi \right]_0^{2\pi} = 0 \quad (\text{for, } l = z) \quad (\text{S19})$$

We then come to the conclusion that:

$$\int_{-\infty}^{\infty} C_x C_y \left( -\frac{5}{2} \mathbf{C} + W^2 \mathbf{C} \right) \Psi^{(0)} d\mathbf{C} \cdot \nabla_{\mathbf{r}} \ln T = 0 \quad (\text{S20})$$

$$\int_{-\infty}^{\infty} C_x C_y \mathbf{C} \Psi^{(0)} d\mathbf{C} \cdot \sum_j \frac{\rho_j \mathbf{F}_j}{\rho_b k_b T} = 0 \quad (\text{S21})$$

Hence, combining Equation (S20) and Equation (S21) together and multiply it by  $-\tau$ , we then get Equation (8).

### 3 Integration in Evaluation of $\eta_d$

Consider the integral in Equation (9),

$$\int_{-\infty}^{\infty} C_x C_y C_x C_y \Psi^{(0)} d\mathbf{C} \quad (\text{S22})$$

In spherical coordinate, this gives us:

$$\int_0^{2\pi} \int_0^{\pi} \int_0^{\infty} C_x C_y C_x C_y C^2 \Psi^{(0)} \sin \theta dC d\theta d\psi \quad (\text{S23})$$

It is known that  $C_x = C \sin \theta \cos \psi$  and  $C_y = C \sin \theta \sin \psi$ . Then, we have:

$$\int_0^{2\pi} \cos^2 \psi \sin^2 \psi d\psi \int_0^{\pi} \sin^5 \theta d\theta \int_0^{\infty} C^6 \Psi^{(0)} dC \quad (\text{S24})$$

Firstly, we consider the integration over  $\psi$ :

$$\cos^2 \psi \sin^2 \psi = \left( \frac{e^{i\psi} + e^{-i\psi}}{2} \right)^2 \left( \frac{e^{i\psi} - e^{-i\psi}}{2i} \right)^2 \quad (\text{S25})$$

$$\int_0^{2\pi} \cos^2 \psi \sin^2 \psi d\psi = -\frac{1}{16} \int_0^{2\pi} [(e^{4i\psi} + e^{-4i\psi}) - 2] d\psi \quad (\text{S26})$$

$$= -\frac{1}{16} \left[ \frac{e^{4i\psi}}{4i} - \frac{e^{-4i\psi}}{4i} - 2\theta \right]_0^{2\pi} \quad (\text{S27})$$

$$= -\frac{1}{16} \left( \frac{2i \sin 4\psi}{2} \right)_0^{2\pi} = \frac{\pi}{4} \quad (\text{S28})$$

where  $i = \sqrt{-1}$ . Secondly, the integration over  $\theta$  is considered:

$$\int_0^\pi \sin^5 \theta d\theta = - \int_0^\pi (1 - \cos^2 \theta)^2 d \cos \theta \quad (\text{S29})$$

$$= - \int_0^\pi (1 - 2 \cos^2 \theta + \cos^4 \theta) d \cos \theta \quad (\text{S30})$$

$$= - \left( \cos \theta \right)_0^\pi + \frac{2}{3} \left( \cos^3 \theta \right)_0^\pi - \frac{1}{5} \left( \cos^5 \theta \right)_0^\pi \quad (\text{S31})$$

$$= -(-1 - 1) + \frac{2}{3}(-1 - 1) - \frac{1}{5}(-1 - 1) = 2 - \frac{4}{3} + \frac{2}{5} \quad (\text{S32})$$

$$= \frac{16}{15} \quad (\text{S33})$$

The integration over  $C$  is evaluated:

$$\rho_b \left( \frac{b}{\pi} \right)^{1.5} \int_0^\infty e^{-bC^2} C^6 dC \quad (\text{S34})$$

$$\int_0^\infty C^6 e^{-bC^2} dC = \int_0^\infty \frac{e^{-br^2}}{2b} 5C^4 dC \quad (\text{S35})$$

$$= \frac{5}{2b} \int_0^\infty \frac{e^{-bC^2}}{2b} 3C^2 dC \quad (\text{S36})$$

$$= \frac{15}{4b^2} \int_0^\infty e^{-bC^2} C^2 dC \quad (\text{S37})$$

$$= \frac{15}{4b^2} \int_0^\infty \frac{e^{-bC^2}}{2b} dC \quad (\text{S38})$$

$$= \frac{15}{8b^3} \cdot \frac{1}{2} \sqrt{\frac{\pi}{b}} = \frac{15}{16b^3} \sqrt{\frac{\pi}{b}} \quad (\text{S39})$$

where  $b = \frac{m}{2k_b T}$ . Finally, we have:

$$\int_{-\infty}^\infty C_x C_y C_x C_y \Psi^{(0)} d\mathbf{C} = \rho_b \frac{4\pi}{15} \left( \frac{b}{\pi} \right)^{1.5} \frac{15}{16b^3} \sqrt{\frac{\pi}{b}} = \frac{\rho_b}{4b^2} = \frac{\rho_b k_b^2 T^2}{m^2} \quad (\text{S40})$$

## 4 Normal Distribution Approximation of $\int_{\phi^+}^\infty P_d(\phi) d\phi$

In our previous work<sup>1,2,3</sup>, the following form of  $P_d(\phi)$  distribution was used:

$$\ln P_d = (N-x) \ln \left[ \left( \frac{1-F}{1-\phi+\lambda} \right)^{1-\phi} \left( \frac{F}{\phi+\lambda} \right)^\phi \right] - \frac{1}{2} \ln \left[ 2\pi(N-x)\phi(1-\phi) + \exp\left(-\frac{\phi^2}{\lambda^2}\right) + \exp\left[-\frac{(\phi-1)^2}{\lambda^2}\right] \right] + \ln(\mu) \quad (\text{S41})$$

where  $\lambda$  is a small number of order of magnitude of  $10^{-5}$ , and  $\mu$  is the normalization constant. Equation (S41) is a result of Stirling approximation. Consider the integral:

$$\int_{\phi^+}^\infty P_d(\phi) d\phi \quad (\text{S42})$$

It can be either approximated as a normal distribution with mean value and covariance being  $F$  and  $F/(N-x)$ , respectively, (cf., Equation (18)) or evaluated numerically based upon the distribution as illustrated in Equation (S41). Figure S1 shows a plot of Equation (S42) as a function of  $N$  computed using the more complicated form in Equation (S41) and the normal distribution approximation in Equation (18) with  $F = 0.45$  and  $x = 2$ , and  $\phi^+ = 0.49$  as well as  $\phi^+ = 0.50$  in the former and latter cases, respectively. The approximation is reasonably good.

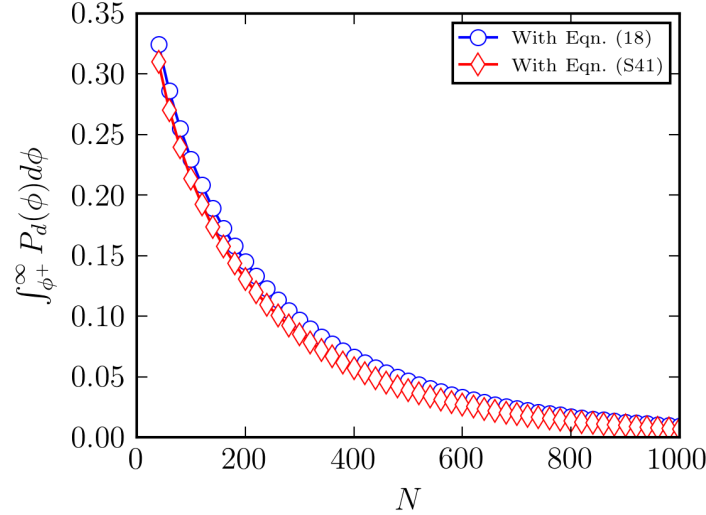


Figure S1: Comparison of  $\int_{\phi^+}^{\infty} P_d(\phi)d\phi$  as a function of  $N$  evaluated using Equation (S41) and Equation (18) with  $F = 0.45$  and  $x = 2$ , as well as  $\phi^+ = 0.49$  and  $\phi^+ = 0.50$  in the former and latter cases, respectively.

## 5 $\hat{\omega}(k)$ of polyethylene with different structures

The total number of  $\hat{\omega}_{\alpha,\gamma}(k)$  of a Gaussian polyethylene is always  $N^2$  regardless of their architectures. If this is not clear, one can write a  $N \times N$  matrix as shown below:

$$\begin{bmatrix} \hat{\omega}_{1,1} & \hat{\omega}_{1,2} & \hat{\omega}_{1,3} & \hat{\omega}_{1,4} & \hat{\omega}_{1,5} \\ \hat{\omega}_{2,1} & \hat{\omega}_{2,2} & \hat{\omega}_{2,3} & \hat{\omega}_{2,4} & \hat{\omega}_{2,5} \\ \hat{\omega}_{3,1} & \hat{\omega}_{3,2} & \hat{\omega}_{3,3} & \hat{\omega}_{3,4} & \hat{\omega}_{3,5} \\ \hat{\omega}_{4,1} & \hat{\omega}_{4,2} & \hat{\omega}_{4,3} & \hat{\omega}_{4,4} & \hat{\omega}_{4,5} \\ \hat{\omega}_{5,1} & \hat{\omega}_{5,2} & \hat{\omega}_{5,3} & \hat{\omega}_{5,4} & \hat{\omega}_{5,5} \end{bmatrix} \quad (\text{S43})$$

where we have let  $N = 5$  as an example. Note that  $\hat{\omega}_{\alpha,\gamma}(k)$  is the Fourier Transform of the probability of finding site  $\gamma$  of the polyethylene with site  $\alpha$  as reference site ( $\omega_{\alpha,\gamma}(r)$ ). We assume that such probability is a normal distribution function of  $r$ . Consider two beads that are separated by one bond in linear and four-arm symmetrical star polyethylene, then  $\omega_{\alpha,\alpha+1}(r)$  is:

$$\omega_{\alpha,\alpha+1}(r) = \left(\frac{b}{\pi}\right)^{1.5} 4\pi e^{-br^2} r^2 \quad (\text{S44})$$

where  $b = \frac{3}{2\langle r^2 \rangle}$ . To evaluate the Fourier Transform of  $\omega_{\alpha, \alpha+1}(r)$ , we have:

$$\frac{1}{k} \left( \frac{b}{\pi} \right)^{1.5} 4\pi \int_0^\infty e^{-br^2} r \left( \frac{e^{ikr} - e^{-ikr}}{2i} \right) dr \quad (\text{S45})$$

where  $k$  is the magnitude of a wavevector and  $i = \sqrt{-1}$ . Then, it can be rewritten:

$$\frac{1}{2ik} \left( \frac{b}{\pi} \right)^{1.5} 4\pi \left[ e^{-\frac{k^2}{4b}} \int_0^\infty e^{-b(r-\frac{ik}{2b})^2} r dr - e^{-\frac{k^2}{4b}} \int_0^\infty e^{-b(r+\frac{ik}{2b})^2} r dr \right] \quad (\text{S46})$$

$$= \frac{1}{2ik} \left( \frac{b}{\pi} \right)^{1.5} 4\pi e^{-\frac{k^2}{4b}} \frac{ik}{2b} \sqrt{\frac{\pi}{b}} \quad (\text{S47})$$

$$= e^{-\frac{k^2}{4b}} \quad (\text{S48})$$

Given the fact that  $b = \frac{3}{2\langle r^2 \rangle}$ , we get:

$$\frac{1}{k} \left( \frac{b}{\pi} \right)^{1.5} 4\pi \int_0^\infty e^{-br^2} r \left( \frac{e^{ikr} - e^{-ikr}}{2i} \right) dr = \exp \left( -\frac{k^2 \langle r^2 \rangle}{6} \right) \quad (\text{S49})$$

And if site  $\alpha$  and site  $\gamma$  is separated by  $n$  bonds, then we have  $\hat{\omega}_{\alpha, \gamma} = \exp(-\frac{nk^2\sigma^2}{6})$ . In ring polymer, it is slightly more tricky that<sup>4,5</sup>:

$$\hat{\omega}_{\alpha, \gamma} = \exp \left[ -\frac{k^2\sigma^2 n(N-n)}{6N} \right] \quad (\text{S50})$$

To see the effect of architecture on the resultant sum  $\hat{\omega}(k)$  more clearly, consider again the case when  $N = 5$ :

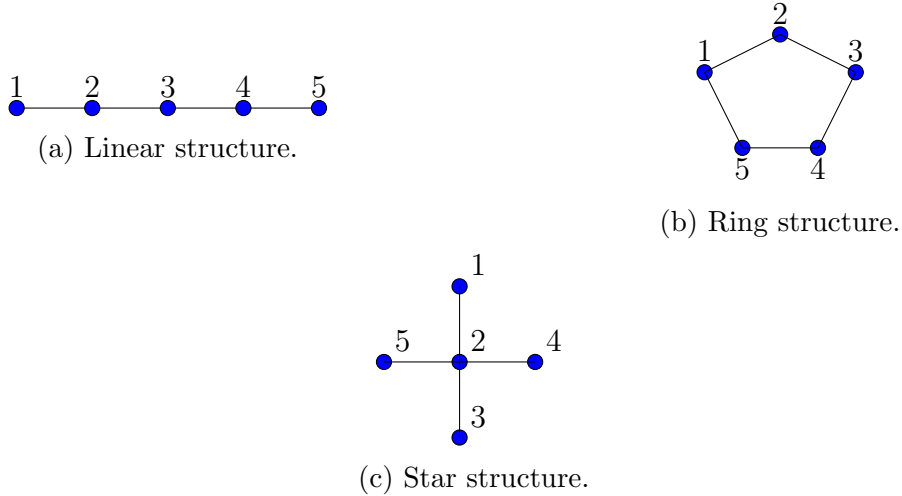


Figure S2: Nomenclature of different bead in polyethylene with different structure.

Let  $f = \exp(-\frac{k^2\sigma^2}{6})$ , then based on the illustration as shown in Figure S2, the corre-

sponding matrices for these three different structures are as below, for linear structure:

$$\begin{bmatrix} 1 & f & f^2 & f^3 & f^4 \\ f & 1 & f & f^2 & f^3 \\ f^2 & f & 1 & f & f^2 \\ f^3 & f^2 & f & 1 & f \\ f^4 & f^3 & f^2 & f & 1 \end{bmatrix} \quad (\text{S51})$$

For ring structure:

$$\begin{bmatrix} 1 & f^{4/5} & f^{6/5} & f^{6/5} & f^{4/5} \\ f^{4/5} & 1 & f^{4/5} & f^{6/5} & f^{6/5} \\ f^{6/5} & f^{4/5} & 1 & f^{4/5} & f^{6/5} \\ f^{6/5} & f^{6/5} & f^{4/5} & 1 & f^{4/5} \\ f^{4/5} & f^{6/5} & f^{6/5} & f^{4/5} & 1 \end{bmatrix} \quad (\text{S52})$$

For four-arm symmetrical star structure:

$$\begin{bmatrix} 1 & f & f^2 & f^2 & f^2 \\ f & 1 & f & f & f \\ f^2 & f & 1 & f^2 & f^2 \\ f^2 & f & f^2 & 1 & f^2 \\ f^2 & f & f^2 & f^2 & 1 \end{bmatrix} \quad (\text{S53})$$

To reiterate,  $\hat{\omega}(k)$  for a particular structure is the summation of all the components in the corresponding matrix.

## 6 Intramolecular Contribution to the Equation of State

To incorporate the intramolecular term in the pressure equation, we firstly have to know the nature of the  $g^{(1)}(r)$ , which must be normalized as follows:

$$\frac{1}{V} \int_0^\infty 4\pi r^2 g^{(1)}(r) dr = 1 \quad (\text{S54})$$

For monoatomic particle,  $g^{(1)}(r) = 1$  as it does not have any internal structure like polymer. Similarly, for a Gaussian polymer, in which  $g^{(1)}(r)$  is the intramolecular radial distribution function per two interacting beads of the same chain. Then, this means that for Gaussian polymer,  $g^{(1)}(r)/V$  must be a normalized probability distribution function  $P(r)$  describing the intramolecular interaction of two beads that  $P(r) = g^{(1)}(r)/V$ . Such probability distribution function for two beads connected with one another in a Gaussian chain is well-known:

$$P(r) = \left( \frac{3}{2\pi \langle r^2 \rangle} \right)^{3/2} \exp \left( - \frac{3r^2}{2 \langle r^2 \rangle} \right) \quad (\text{S55})$$

$\langle r^2 \rangle$  is the mean square statistical step length of the Gaussian chain. Now, we consider the harmonic bond stretching potential  $u_{bond}$ .

$$\frac{du_{bond}}{dV} = \frac{du_{bond}}{dr} \frac{dr}{dV} \quad (\text{S56})$$

$r$ , which is the distance between two beads interacting with one another by  $u_{bond}(r)$ , can be expressed as follows:

$$r = \sqrt{V^{2/3}(x'^2 + y'^2 + z'^2)} \quad (\text{S57})$$

Then,

$$\frac{dr}{dV} = \frac{r}{3V} \quad (\text{S58})$$

Therefore:

$$\frac{du_{bond}}{dV} = \frac{du_{bond}}{dr} \frac{dr}{dV} = \frac{r}{3V} \frac{du_{bond}}{dr} \quad (\text{S59})$$

The internal energy of one Gaussian chain with only intramolecular interaction is:

$$U_N = \sum_{\alpha=1}^{N-1} u_{bond}(\mathbf{r}_\alpha, \mathbf{r}_{\alpha+1}) \quad (\text{S60})$$

In statistical mechanics, the pressure of one chain with only intramolecular interaction,  $P_1$  can be expressed as:

$$P_1\beta = \frac{1}{Z} \left( \frac{\partial Z}{\partial V} \right)_{N,T} \quad (\text{S61})$$

where  $Z = \int_0^\infty e^{-\beta U_N} d\mathbf{r}_1 d\mathbf{r}_2 \dots d\mathbf{r}_N$  if we only consider the intramolecular harmonic bond stretching interaction. Then, we have:

$$\left( \frac{\partial Z}{\partial V} \right)_{N,T} = - \int_0^\infty e^{-\beta U_N} \beta \frac{dU_N}{dV} d\mathbf{r}_1 d\mathbf{r}_2 \dots d\mathbf{r}_N \quad (\text{S62})$$

It is known that:

$$\frac{dU_N}{dV} = N \frac{d}{dV} u_{bond}(\mathbf{r}_1, \mathbf{r}_2) \quad (\text{S63})$$

This is because all the bonds are identical, and  $u_{bond}(r)$  is a type of two-body interaction. In addition, by definition, for a single chain, we have:

$$g^{(1)}(\mathbf{r}_1, \mathbf{r}_2) = \frac{\rho^{-1} V \int_0^\infty \dots \int_0^\infty e^{-\beta U_N} d\mathbf{r}_3 d\mathbf{r}_4 \dots d\mathbf{r}_N}{Z} \quad (\text{S64})$$

Therefore, we can rewrite Equation (S62) in spherical coordinates:

$$\frac{1}{Z} \left( \frac{\partial Z}{\partial V} \right)_{N,T} = -4\pi N\rho \int_0^\infty g^{(1)}(r) \beta r^2 \frac{du_{bond}}{dV} dr = -\frac{4\pi N\rho}{3V} \int_0^\infty g^{(1)}(r) \beta r^3 \frac{du_{bond}}{dr} dr \quad (\text{S65})$$

Note that  $\rho_b = N\rho$ . Then we have:

$$P_1 = -\frac{4\pi\rho_b}{3V} \int_0^\infty g^{(1)}(r) r^3 \frac{du_{bond}}{dr} dr \quad (\text{S66})$$

Honnell *et al.*<sup>6</sup> also demonstrated similar results. We then obtained Equation (30) by the fact that  $P(r) = g^{(1)}(r)/V$ .



## References

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