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Single long-polymer translocation through a long pore*

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This paper theoretically studies the free energy and conformational entropy of a long polymer threading a long nanopore ($n_0/N \geq 0.1$) on external electric field. The polymer expanded model is built in this paper, that is, a single long polymer chain with N monomers (each of size a) threading a pore with n_0 monomers can be regarded as polymer with $N + n_0$ monomers translocating a 2-dimension hole embedded in membrane. A theoretical approach is presented which explicitly takes into account the nucleation theory. Our calculations imply that, the structure of polymer changes more acutely than other situation, while its leading monomer reaches the second vacuum and its end monomer escapes the first vacuum. And it is also shown that the length scale of polymer and pore play a very important role for polymer translocation dynamics. The present model predicts that the translocation time depends on the chemical potential gradient and the property of the solvent on sides of pore to some extent.

Keywords: polymer expansion, nucleation theory, conformational entropy, translocation time
PACC: 0520D, 8715H, 8715B

1. Introduction

Translocation of polymers across a narrow pore is an event which is important to many biological systems. Examples include the injection of viral DNA into a host cell, the motion of DNA and RNA molecules across nuclear pores, DNA packaging into viral capsids, gene swapping through bacterial pili, translocation of protein from *cis* to *trans* side of a membrane through channels, and involve the motion of biopolymers across membranes.[1−2] Accordingly, the mechanisms of polymer translocation have become a highlight of numerous experiments.[3−10]

Although the actual translocation is very complicated due to many factors, a simple model was built to describe a polymer chain made up of N monomers translocating through a pore, which may provide useful insights for understanding basic features of a polymer threading membrane. Several theoretical models have been developed in order to explain these experimental findings. Sung and Park utilized Fokker–Plank equation to study the Gaussian chain translocation dynamics through a pore embedded in a flat membrane where the monomer friction across the pore is proportional to the length scaling of the polymer.[11−15] Muthukumar dealt this process with nucleation theory.[16−18] Oster group used modified one dimensional drift diffusion equation to build a Brownian ratchet mechanism for protein translocation.[19,20] Lubensky and Nelson also studied 1-dimensional drift diffusion equations incorporating a simple model of monomer-pore interaction.[21] Di Marzio and co-workers studied thermodynamic phase transitions for an isolated polymer molecule in the limitation of infinite molecular weight.[22−24] Pu and Smith carried out Brownian dynamics simulation study of the translocation of single polymer chain across a nanopore.[25] Chen used off-lattice Monte Carlo simulations to study the driven translocation dynamics of a polynucleotide chain through a nanopore.[26] Matsuyama studied theoretically the phase transitions of a polymer escaping from a pore based on the Flory theory of the coil-globule transitions of a single polymer chain.[27−29] Ambjörnsson *et al* took into account the nanopore length and studied both the polymer entrance into the pore and the

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translocation process.[30] Sebastian and Paul viewed the polymer translocation as the motion of a kink, which travelled in the direction opposite to polymer transport.[31] Chern *et al*'s 3-dimensional model illustrated that it is indeed impossible to capture many essential feature of the translocation process by using an appropriate 1-dimension model.[32] And Xie *et al*[33] used metropolis molecular dynamics simulation[34,35] combined with bond-fluctuation method to study the dynamic process of single DNA chain through an entropic trapping channel driven by electric field. All these theoretical works provided reasonable descriptions of polymer threading through the nanopore for very large polymers. Although Refs.[11, 16, 30] mentioned that the moving polymer molecule should overcome the entropic barrier, these theories were paid less attentions to entropy than the conformational scaling of the polymer.

From the empirical data, it is very clear that the typical length scale of the pores and translocating polymers are of ten nanometres and the polymer is subjected to significant conformational changes during threading.[2,31] Modern polymer physics has achieved great success with models in which the polymer is regarded as a flexible, uniform 'string' whose conformational entropy dominates the system's behaviour.[32,36] Therefore, it has to effectively apply entropy (polymer and statistical physics) at molecular level to understand the above mentioned translocation processes.

In this paper, we overcame the fault of the previous theories, which did not consider the realistic length of pore, and considered that the length scaling of the polymer and pore are large enough to satisfy the current experimental condition. The translocation pore can be regarded as a long pore when the ratio between the pore and polymer exceeds 10%. Based on the partition function,[37] the classic nucleation theory[16,17,29] and our polymer expanded model, we considered the threading dynamics of a polymer which passes through a pore imbedded in a membrane, and numerically calculated the free energy, the entropy and the translocation time.

2. Properties of polymer translocation

The monomers of the polymer on one side of the membrane can translocate into the other side only by passing through the pore whose diameter is comparable with the size of the chemical repeat unit on the polymer[29] under the influence of an external electric field. Suppose a single long polymer chain with N monomers (each of size a) moving from region I to region III through a membrane pore of length $l_0(= n_0 a)$ and diameter D, as shown in Fig.1, where a is the length of each monomer. In realistic experiments,[7] a, n_0 are equal to 4Å and 12 respectively. In the thermal equilibrium state, the monomer segments are divided into region I, II and III. The conformational entropy of the polymer was derived from the classic nucleation theory for polymer solution.[17]

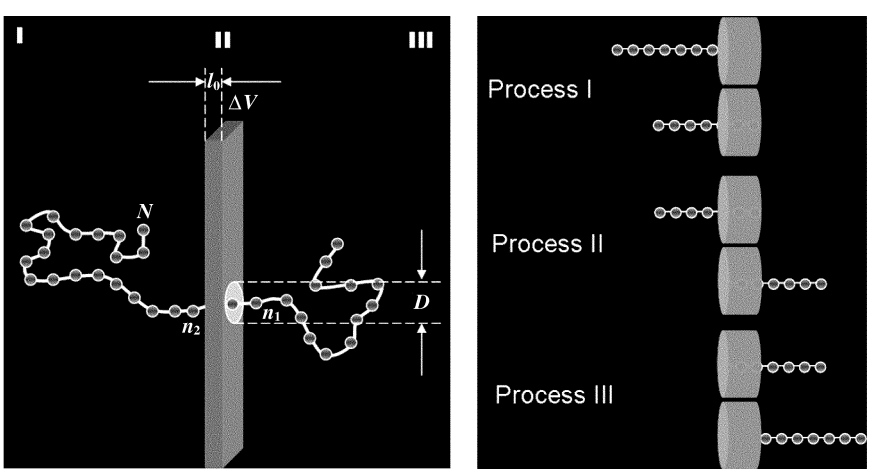


Fig.1. Schematic representation of a N-monomer polymer in the process of translocation through a cylindrical nanopore of length l_0 and diameter D. n is the number of monomers on one side and n_0 is the number of monomers on the nanopore. ΔV is potential energy (about 50–200mV) in realistic experimental condition. The right picture shows that the whole process was divided into three parts.

Under the experimental condition, it is impossible for the monomer to return when it enters into the pore since the energy gained by a single monomer for translocating the membrane pore is much higher than its thermal energy. We simply assume that the interaction between the polymer and the pore can be ignored and chemical potential inside the pore is uniform. Let the chemical potential of the monomer in region I, II, III be ε_1, ε_2, and ε_3 respectively (see Fig.1). In addition, the polymer cannot be torn apart for typical experimental voltages. In this paper, the process of translocation is assumed to start as soon as the leading monomer enters the pore and to end as soon as the end monomer leaves the pore, and the whole process can be divided into process I, process II and process III (see Fig.1.). So, N monomers' polymer threading n_0 monomers' pore can be regarded as $N+n_0$ monomers' polymer translocating a 2-dimensional hole embedded in membrane in order to ensure the whole polymer translocation continuity, which is named polymer expanded model. We also assume that the length of polymer is much larger than the pore's, that is, $N > n_0$.

Consider the entropy landscape associated with a translocation of a polymer of N monomer from region I to III. The entropy of the polymer chain is given by

$$S = S_{\mathrm{I}} + S_{\mathrm{II}} + S_{\mathrm{III}}. \tag{1}$$

The relation between the entropy and free energy of the polymer chain is given by

$$F = U - TS, \tag{2}$$

the free energy of the polymer chain can be expressed by

$$F = F_{\mathrm{I}} + F_{\mathrm{II}} + F_{\mathrm{III}}. \tag{3}$$

Where F_{I}, F_{II}, F_{III} is the free energy of the polymer in the region I, II and III, respectively. Based on the partition function[37] and our analysis, we get the overall free energy of polymer

$$F_{n,\mathrm{I}} = (1-\gamma_1')\beta^{-1}\ln(N-n) + n\Delta\varepsilon_{21}, \quad (0 \le n \le n_0), \tag{4}$$

$$F_{n,\mathrm{II}} = (1-\gamma_1')\beta^{-1}\ln(N-n) + (1-\gamma_2')\beta^{-1}\ln(n-n_0) + (n-n_0)\Delta\varepsilon_{31}, \quad (n_0 < n \le N-1), \tag{5}$$

$$F_{n,\mathrm{III}} = (1-\gamma_2')\beta^{-1}\ln(n-n_0) + (n-n_0)\Delta\varepsilon_{32}, \quad (N \le n \le N+n_0). \tag{6}$$

The first or first-second term is the entropic contribution. The last term represents the energy gained by moving n monomers into the pore including the effects of the external field and chemical changes. $\beta^{-1} = k_{\mathrm{B}}T$, k_{B} is Boltzmann constant, T is absolute temperature. γ_1' is a parameter describing the properties of end-anchored polymer chain in region I and which is equal to 0.5, 0.69, and 1.0 for Gaussian, self-avoiding, and rodlike chains respectively.[37] And γ_2' describes the properties of the polymers in the region III. The chemical potential differences $\Delta\varepsilon_{21}=\varepsilon_2-\varepsilon_1$, $\Delta\varepsilon_{31}=\varepsilon_3-\varepsilon_1$ and $\Delta\varepsilon_{32}=\varepsilon_3-\varepsilon_2$ are the chemical potential differences per monomer that will depend on the type of monomer, and n varies between 0 and $N+n_0$. In fact, F exhibits a free energy barrier of the polymer, which depends on the conformational statistics of polymer chain and the values of the chemical potential ε_1, ε_2 and ε_3. The overall free energy is not a continuous function in the parameter n. Its result is shown in Fig.2. Our assumption: N=100, n_0=12 for

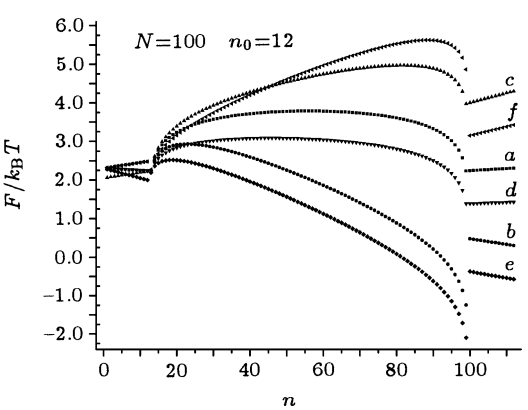


Fig.2. Plot of F in unit of β^{-1} against n. *a* $\gamma_1'=\gamma_2'=0.5$, $\Delta\varepsilon_{21} = \Delta\varepsilon_{31} = \Delta\varepsilon_{32}=0$; *b* $\gamma_1'=\gamma_2'=0.5$, $N\beta\Delta\varepsilon_{21}= N\beta\Delta\varepsilon_{32}=-2$, $N\beta\Delta\varepsilon_{31}=-4$; *c* $\gamma_1'=\gamma_2'=0.5$, $N\beta\Delta\varepsilon_{21}= N\beta\Delta\varepsilon_{32}=2$, $N\beta\Delta\varepsilon_{31}=4$; *d* $\gamma_1'=0.5$, $\gamma_2'=0.69$, $\Delta\varepsilon_{21} = \Delta\varepsilon_{21} = \Delta\varepsilon_{21}=0$; *e* $\gamma_1'=0.5$, $\gamma_2'=0.69$, $N\beta\Delta\varepsilon_{21}= N\beta\Delta\varepsilon_{32}=-2$, $N\beta\Delta\varepsilon_{31} = -4$; *f* $\gamma_1'=0.5$, $\gamma_2'=0.69$, $N\beta\Delta\varepsilon_{21}= N\beta\Delta\varepsilon_{32}=2$, $N\beta\Delta\varepsilon_{31}=4$.

free energy and entropy's calculation and N=1000, n_0=120 for translocation time. When $\Delta\varepsilon_{21} = \Delta\varepsilon_{31} = \Delta\varepsilon_{32}$=0, $\gamma_1' = \gamma_2' = 0.5$, the free energy is symmetric in whole polymer translocation process, see the curve a of Fig.2. But, the barrier becomes asymmetric for $\gamma_1' \neq \gamma_2'$ even if $\Delta\varepsilon_{21} = \Delta\varepsilon_{31} = \Delta\varepsilon_{32}$=0, see the curve d of Fig.2. The chemical potential difference is a key factor for determining the tendencies of polymer free energy. This is illustrated in Fig.2. When the leading monomer reaches the region III and the end monomer escapes the region I, the free energy of polymer has a transition, which illustrates that the structure of polymer has a large transformation. The following entropy calculation also testifies this fact. Based on the above equations and ignoring the unnecessary constant terms, we can express the entropy of the polymer as

$$S = \begin{cases} (1-\gamma_1')k_\beta \ln(N-n), & n \le n_0, \\ (1-\gamma_1')k_\beta \ln(N-n) + (1-\gamma_2')k_\beta \ln(n-n_0), & n_0 < n \le N-1 \\ (1-\gamma_3')k_\beta \ln(n-n_0), & N \le n < N+n_0. \end{cases} \tag{7}$$

The overall entropy is also a non-continuous function in the parameter n in translocation, but it doesn't depend on the chemical potential mismatch, which is different from the free energy. From this equation, the configuration entropy is dependent of n. and its differential relation can be given by

$$\frac{\partial S}{\partial n} = \begin{cases} \dfrac{(\gamma_1'-1)k_\beta}{N-n}, & n \le n_0, \\ \dfrac{(\gamma_1'-1)k_\beta}{N-n} + \dfrac{1-\gamma_2')k_\beta}{n-n_0}, & n_0 < n \le N-1, \\ \dfrac{(1-\gamma_2')k_\beta}{n-n_0}, & N \le n < N+n_0. \end{cases} \tag{8}$$

These results are illustrated in Fig.3. When γ_1'=γ_2' =0.5, the entropy is symmetric in whole polymer translocation process, see the curve a of Fig.3. But the barrier becomes asymmetric for $\gamma_1' \neq \gamma_2'$, see the curve b of Fig.3. The differential relation between the entropy S and the number of monomer n is symmetric for given n=56 whenever γ_1'=γ_2' or $\gamma_1' \neq \gamma_2'$. And entropy of polymer translocation is also sustained

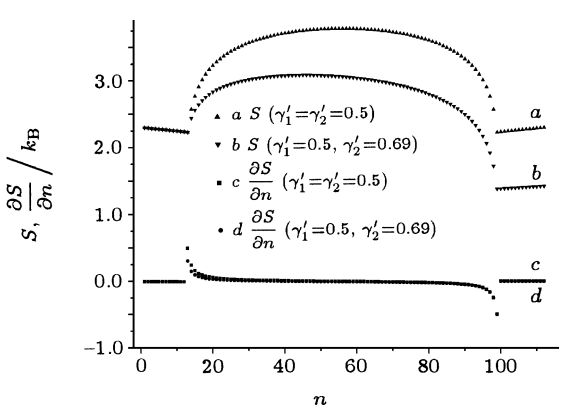


Fig.3. Plot of S and $\partial S/\partial n$ in unit of k_B against n. *a* S: N=100, n_0=12, γ_1'=γ_2' =0.5; *b* S: N=100, n_0=12, γ_1'=0.5, γ_2'=0.69; *c* $\partial S/\partial n$: N=100, n_0=12, γ_1'=γ_2'=0.5; *d* $\partial S/\partial n$: N=100, n_0=12, γ_1'=0.5, γ_2'=0.69.

that the structure of polymer has a large transformation when the leading monomer reaches the region III and the end monomer escapes the region I.

The mean translocation time was usually measured in previous experiments,[3−10] so it is necessary for us to evaluate it. Following the nucleation theory,[38] the transport of the polymer across the nanopore can be described by a Master equation[16]

$$\frac{\partial}{\partial t}P_n(i,t) = k_{n-1}P_{n-1}(i,t) - k_n'P_n(i,t) - k_nP_n(i,t) + k_{n+1}'P_{n+1}(i,t). \tag{9}$$

Where $P_n(i,t)$ is the probability of emerging a "core" of n segments in region i=I, II or III at time t, k_n is the rate constant associated with the local friction in transfer of one monomer through the hole so that a nucleus of n monomers grows into $n+1$ monomers, and k_n' is the rate constant of removing one monomer from the n monomers. Using the detailed balance, we can express k_{n+1}' in terms of k_n,

$$k_{n+1}' = k_n e^{\beta(F_{n+1}-F_n)}. \tag{10}$$

Where k_{n+1}' and k_n are also assumed to be independent of n for the previous researches[16] since we deal

with only homopolymer, and they are taken to be a non-universal constant K_0^{-1}. Using Eq.(10) and continuum Smoluchovskii equation,[36] we obtain

$$\frac{\partial}{\partial t}P_n(i,t) = \frac{\partial}{\partial n}\left[k_n\beta\frac{\partial F_{n,i}}{\partial n}P_n(i,t) + k_n\frac{\partial}{\partial n}P_n(i,t)\right], \quad (11)$$

where the first term is polymer drift contribution. When the first term in (11) dominates the situation, the mean translocation time τ can be driven as a sum of mean first-passage times in each region, that is, $\tau=\tau_{\rm I}+\tau_{\rm II}+\tau_{\rm III}$. $\tau_{\rm I}$, $\tau_{\rm II}$ and $\tau_{\rm III}$ is mean first-passage times in region I, II, III respectively, which can be given as

$$\tau_{\rm I} = K_0\int_0^{n_0} {\rm d}m\exp(\beta F_{m,{\rm I}})\int_0^m {\rm d}n\exp(-\beta F_{n,{\rm I}}), \quad (12)$$

$$\tau_{\rm II} = K_0\int_{n_0}^{N} {\rm d}m\exp(\beta F_{m,{\rm II}})\int_0^m {\rm d}n\exp(-\beta F_{n,{\rm II}}), \quad (13)$$

$$\tau_{\rm III} = K_0\int_{N}^{N+n_0} {\rm d}m\exp(\beta F_{m,{\rm III}}) \times\int_0^m {\rm d}n\exp(-\beta F_{n,{\rm III}}). \quad (14)$$

Here $K_0^{-1} = k_n$ has been mentioned in previous paragraph. As is clearly seen, from the Ref.[16,17], the potential chemical difference controls the polymer translocation time τ.

For simplicity, if the chemical potential gradient is absent, that is, $\Delta\varepsilon_{21} = \Delta\varepsilon_{31} = \Delta\varepsilon_{32}$=0, the exactly expression of the translocation time τ ($=\tau_{\rm I}+\tau_{\rm II}+\tau_{\rm III}$) can be obtained,

$$\tau = \frac{K_0(N+n_0)^2}{\gamma_1'}\left[\frac{\left(\frac{N}{N+n_0}\right)^{\gamma_1'} - \left(\frac{N}{N+n_0}\right)^2}{(2-\gamma_1')\left(\frac{N}{N+n_0}\right)^{\gamma_1'}} - \frac{1-\left(\frac{N}{N+n_0}\right)^2}{2}\right] + aK_0N^2 + \frac{K_0(N+n_0)^2}{2-\gamma_2'}\left[\frac{1-\left(\frac{N}{N+n_0}\right)^{\gamma_2'}}{\gamma_2'} - \frac{1-\left(\frac{N}{N+n_0}\right)^2}{2}\right]. \quad (15)$$

Here, a is a constant depending on γ_1' and γ_2', which is equal to 0.5 for the special case of γ_1'=γ_2'=0.5. Figure 4 illustrates the translocation time for different pore length. When $N >> n_0$, the translocation time is governed by the process II (see Fig.4(a)). The process I, II is determined for long pore (n_0/N$\geq$0.1) (see Fig.4(b)). For given polymer, the translocation time in process II is similar when the length of pore is not considered. So, we only need to compare the translocation time in region I, III for different pore length. Figure 5(a) shows that it is more difficult for polymer to enter long pore than enter short pore. And Fig.5(b) illustrates that, for given long pore, the longer the polymer is, the shorter the translocation time of process III is.

Without considering the length scaling of the pore, that is, n_0=0, the translocation time τ equals to aK_0N^2 which is in agreement with the previous results.[16] For $N >> n_0$, using Taylor series and neglecting the higher terms, we can rewrite the overall translocation time τ as

$$\tau = K_0n_0^2 + aK_0N^2. \quad (16)$$

Because of $N \sim n_0$, τ can be expressed as

$$\tau = 2K_0N^2\left(\frac{1}{2-\gamma_1'} + \frac{1}{\gamma_2'} + \frac{a_0}{2}\right). \quad (17)$$

In these conditions the translocation time τ is approximately proportional to N^2.

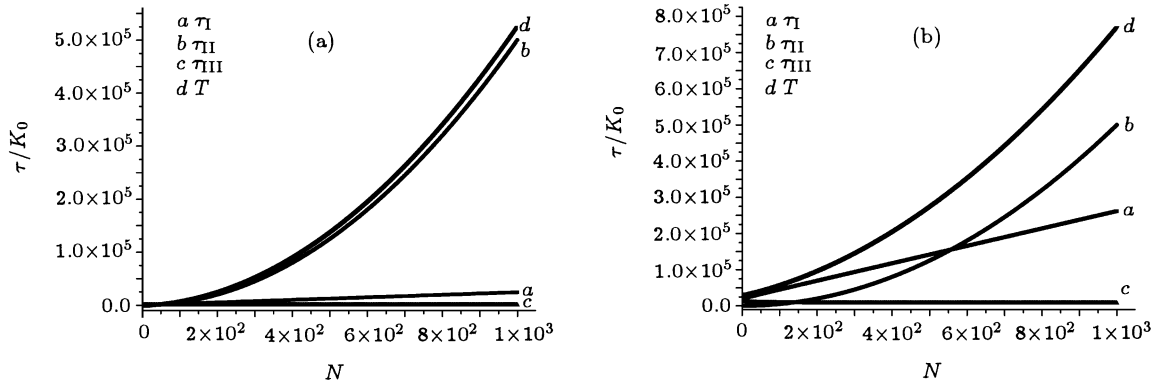


Fig.4. Translocation time τ in unit of K_0 against N. $\gamma_1'=\gamma_2'$=0.5 (a) n_0=12; (b) n_0=120.

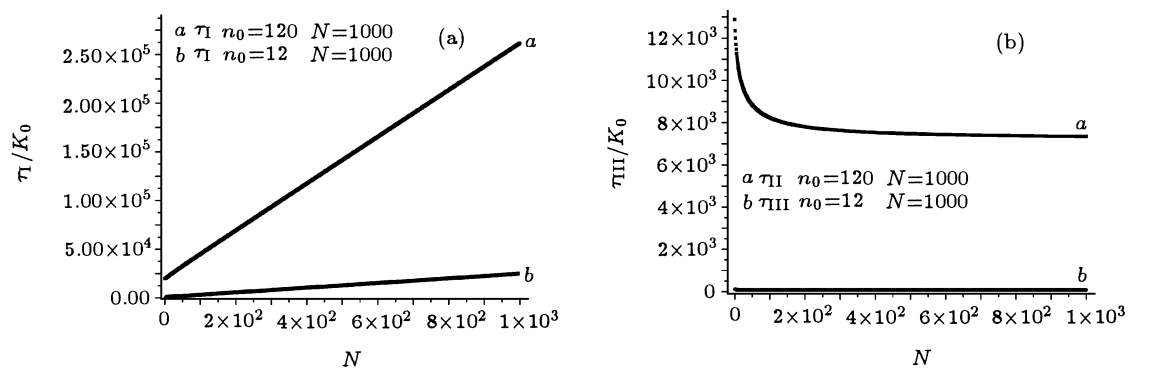


Fig.5. Comparison the translocation time in region I, III for different length pore $\gamma_1'=\gamma_2'$=0.5 (a) n_0=12 (b) n_0=120.

For realistic condition, if the entropic terms in Eqs.(4), (5) and (6) are weak in comparison with the $\Delta\varepsilon$ terms, and $|\Delta\varepsilon_{21}| > 1/n_0$, $|\Delta\varepsilon_{31}| > 1/(N-n_0)$, $|\Delta\varepsilon_{32}| > 1/n_0$, the translocation time τ along the favourable chemical potential gradient ($\Delta\varepsilon < 0$) is obtained by

$$\tau = K_0\beta^{-1}\left(\frac{n_0}{|\Delta\varepsilon_{21}|}+\frac{N}{|\Delta\varepsilon_{31}|}+\frac{n_0}{|\Delta\varepsilon_{32}|}\right). \qquad (18)$$

It shows that the translocation time τ is governed by the second term and is approximately proportional to N if $N >> n_0$. And it is clearly seen that the chemical potential difference between the regions I and II also controls the time in this condition. But, when $|\Delta\varepsilon_{21}| < 1/n_0$, $|\Delta\varepsilon_{31}| < 1/(N-n_0)$, $|\Delta\varepsilon_{32}| < 1/n_0$, the time τ can be expressed as

$$\tau = \frac{1}{2}K_0N^2 + K_0n_0^2. \qquad (19)$$

Which denotes the translocation time τ is approximately proportional to N^2 and is independent of the property of solvent in each side of the pore when $N >> n_0$. But if $N \sim n_0$, the length scaling l_0, i.e. n_0 will also govern the overall translocation time.

3. Conclusions

We have derived explicit expression for the free energy and its entropy in different regions of polymer translocation across a pore without considering the interaction between the polymer and the pore and the fluctuation of the chemical potential inside the pore. The overall translocation time τ has also been derived based on polymer expanded model. We found that the length scaling of polymer and pore is a crucial factor for dynamics of polymer translocation. The overall free energy and entropy of polymer are not continuous functions in the process of the polymer translocation. Both of them have transition when the leading monomer reaches the region III and the end monomer escapes from the region I, which supports that the structure of polymer has an acute transformation in these two situations. When $N >> n_0$ and $N \sim n_0$, the translocation time τ is approximately proportional to N^2 with different coefficient for $\Delta\varepsilon_{21}=\Delta\varepsilon_{31}=\Delta\varepsilon_{32}$=0, which is in agreement with the previous research results. But if the entropy terms in the free energy equation are weak

in comparison with the $\Delta\varepsilon$ terms, for $|\Delta\varepsilon_{21}| > 1/n_0$, $|\Delta\varepsilon_{31}| > 1/(N - n_0)$, $|\Delta\varepsilon_{32}| > 1/n_0$, $\tau \sim N$ and for $|\Delta\varepsilon_{21}| < 1/n_0$, $|\Delta\varepsilon_{31}| < 1/(N - n_0)$, $|\Delta\varepsilon_{32}| < 1/n_0$, $\tau \sim N^2$. All these show that the polymer translocation is independent of the solvent condition and the potential chemical difference.

Interaction between the polymer and the pore, temperature of the solvent, not considered in this paper, are also likely to play an important role. We hope that our results encourage further experimental and analytical studies of forced translocation including these factors. In particular, it would be interesting to better characterize the manner in which the leading monomer reaches the region III and the end of the monomer escapes from the region I.

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