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Factors Affecting Polymer Translocation Through a Nanopore in a Membrane

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Monte Carlo simulations were used to study the translocation of a flexible polymer through a pore in a membrane, assuming an attractive interaction between the monomers and the membrane on the *trans* side of the membrane and no interaction on the *cis* side. For the case $T < T_c$ (the temperature corresponding to the minimum in the translocation time τ), the value of τ decreases with increasing temperature, whereas for $T > T_c$, τ increases with increasing temperature. The translocation time depends on the absorbed energy u_0 in a nontrivial way. The value of τ increases initially upon increasing u_0 before it begins to decrease. The variation of the translocation time with respect to the solvent quality was also studied. It showed that there is a transition, as the solvent quality improves from “poor” to “good”: when $\varepsilon_{AB} < \varepsilon_c$ (the interaction energy corresponding to the minimum in τ), τ decreases with increasing the value of ε_{AB} ; when $\varepsilon_{AB} > \varepsilon_c$, τ increases with increasing ε_{AB} . When the chain length was changed, it was found that when the absorbed energy u_0 was greater than u_c , τ was proportional to $N^{1.602}$; for $u_0 < u_c$, $\tau \propto N^{2.248}$. As the solvent quality improved from “poor” to “good,” the translocation probability increased initially before becoming stable.

Key words: Monte Carlo simulation, Translocation time, Absorbed energy, Solvent quality

I. INTRODUCTION

The translocation of polymers through narrow pores is an important phenomenon in many biological systems, such as the translocation of RNA through nuclear pores [1] and the incorporation of proteins into membranes in nearly all cells [2]. *In vitro* experiments have demonstrated that single-strand DNA polymers can be driven through a single α -hemolysin pore (inside diameter: ca. 2 nm) under the influence of an external field [3,4]. Interest in this area has been boosted because of the possible use of biomolecules in, among other things, drug delivery and gene transfer [5-8]. In attempts to understand the translocation process, many theoretical and computational studies of polymer translocation have been undertaken [9-20]. These studies have, in the main, been restricted to considerations of one-dimensional diffusion processes [9] and the nucleation model [11]. Sung and Park studied the transport of a Gaussian chain under the special conditions that the segment friction across the pore was proportional to the polymer length [9]. Muthukumar treated the process using a classical nucleation theory in which the monomer friction was assumed to be dictated by the ratchet potential associated with the pore [11]. Because polymer translocation is such a ubiquitous phenomenon, it may not necessarily be the case that a single universal mechanism operates in all circumstances; hence, it remains worthwhile to perform more-detailed

studies of the various mechanisms.

It is well known that the conformation of a polymer chain depends on the solvent, the temperature, and the ionic strength of the medium [21-24]. In a “good” solvent, the polymer will exist in a coiled state in which its high polymer chain entropy will have a great effect on the progress of polymer translocation through the pore; in a “poor” solvent, however, the polymer will have a globular conformation, such that a pulling force must be applied to initially unwind the polymer prior to its being dragged through the pore. Previous studies [9-20,25-30] have rarely paid any attention to the effects of the “poor” solvent. In this work, we consider the process of polymer translocation through a pore in solvents ranging from “poor” to “good”.

The details of the model constructed to perform the simulations of polymer translocation and the simulation techniques are outlined in the following section. The considerations of homopolymer chains that translocate through a membrane were limited. The translocation time was studied as a function of the simulation temperature, the solvent (ranging from “poor” to “good”), and the strength of the external field on the *trans* side. The dependence of the translocation time and velocity on the length of the polymer was also addressed. Finally, the dependence of that translocation probability on the solvent quality and on the value of N_{trans} , i.e., the number of monomers that exist on the *trans* side of the membrane, were studied. The results are discussed in comparison to other simulation studies and experimental observations.

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II. MODEL AND SIMULATION

For many problems in polymer science, a detailed description of the chemical structure of the polymer is not necessary: it suffices to focus on a coarse-grained model of a polymer chain that maintains only the essential features of the macromolecule, such as the connectivity of the chain, its flexibility, and the extent of nonbonded (excluded volume) interactions. In these present Monte Carlo simulations, the single-site bond fluctuation model was used. The evolution of the chain configuration was achieved by random displacement of a single unit to its eighteen neighboring sites in three dimensions. The distance between a single unit and its nearest neighboring sites was restricted to within 1 and $\sqrt{2}$. Although each attempted move might have changed the bond length, the chain connectivity was restricted to a distance between 1 and $\sqrt{2}$. No bond crossing was allowed. If one attempted move violated the excluded volume, bond crossing, or bond length restrictions, it was rejected. The acceptance or rejection of an attempted move that satisfied both the excluded volume and the bond length conditions was further governed by the Metropolis rule [31], namely, it was accepted if the energy change, ΔE , was negative. Otherwise it was accepted with the probability P determined as follows:

$$P = \exp(-\Delta E/k_B T) \quad (1)$$

$$\Delta E = \Delta N_{AA}\varepsilon_{AA} + \Delta N_{BB}\varepsilon_{BB} + \Delta N_{AB}\varepsilon_{AB} + \Delta V(r) \quad (2)$$

where ΔE is the energy change of movement, ΔN is the difference between the number of the nearest-neighboring pairs of the sites occupied by monomers or by solvents before and after movement, ε is the reduced interaction energy gained after the two neighboring sites were occupied by monomers, solvents, or membranes, and the subscripts A and B denote monomers and solvents units, respectively. The term $\Delta V(r)$ is the difference between the interaction potential between a monomer and membrane before and after movement. k_B is the Boltzmann constant and T is the absolute temperature. The interaction potential between a monomer and membrane is defined using Eq.(3). In this simulation, the following condition $\varepsilon_{AA}=\varepsilon_{BB}=0$ was set. Following the approach taken by Park and Sung [19], it considered an asymmetric membrane for which the polymer segments interact with the membrane attractively on the *trans* side, but on the *cis* side recognize the membrane only as an impenetrable wall. In the x - y plane the membrane is infinite. But in the z direction, the thickness of the membrane (W) was 1. The pore that exists in the flat membrane is sufficiently small that the double threading does not occur. The interaction potential between a monomer and the membrane is defined as follows:

$$\begin{aligned} V(r) &= -u_0, & 0 < z \leq a \\ V(r) &= 0, & z \leq 0 \\ V(r) &= 0, & z > 0 \end{aligned} \quad (3)$$

where $u_0 > 0$ is the strength of the absorbed energy and a is the range of attraction. The system of the polymer translocation through a nanopore in a membrane is initialized that N_{trans} monomers are on the *trans* side of the membrane, where the attractive potential acts, while $N - N_{trans}$ monomers are present on the *cis* side [19]. In this simulation, we applied 10^7 Monte Carlo steps (MCS) to equilibrate the initial conformation of the polymer.

III. RESULTS OF SIMULATION

A. Variation of τ with temperature

The effect of temperature on the translocation time of a polymer chain through a nanopore under “good” solvent conditions has been studied extensively [20,32]. In comparison, the situation in a kind of “poor” solvent has rarely been noted, even though it is always encountered in practical applications. Thus, our focus in this simulation is on the translocation behavior of polymer chains in a “poor” solvent. A polymer chain of length $N=40$ was considered which has a reduced interaction energy between the chain monomer and solvent of $\varepsilon_{AB}=0.1$. The strength of absorbed energy was set at either $u_0=1.0$ or 0.6 . The value of N_{trans} was set at zero. In Fig.1, the translocation time distributions are plotted as histograms. It was found that the distribution of translocation time displays non-Gaussian characteristics. This finding is similar to the experimental results obtained for the translocation time distribution of DNA across a protein channel [3], in which the salient feature is the strong non-Gaussian characteristics of the translocation time distribution. The non-Gaussian characteristic feature was also predicted by the 1D diffusion model [12,30]. Figure 2 presents the translocation time of the polymer chain plotted against reduced temperature. Two distinct regions were observed: in the region $T < T_c$ (where T_c is the temperature corresponding to the minimum value of τ), the translocation time τ decreases upon increasing the temperature, but it increases in the region $T > T_c$. To understand these observations, we must consider that two components exist that present barriers to the pulling force arising from the potential difference across the membrane [33]. The first one, the cohesive energy barrier (E_c), is due to the intermonomer potential, which makes the polymer favor adopting a compact conformation. If the polymer exists in such a conformation, a pulling force must be applied initially to unwind the polymer (i.e., to break the intermonomer interactions) prior to its being dragged through the pore. The other, the entropy barrier (E_s), arises from the temperature fluctuations. In general, the entropy barrier E_s increases and the cohesive energy barrier E_c decreases upon increasing the temperature. These two contrasting effects result in a minimum net barrier to pulling at a critical temperature and, correspondingly, a minimum translocation time.

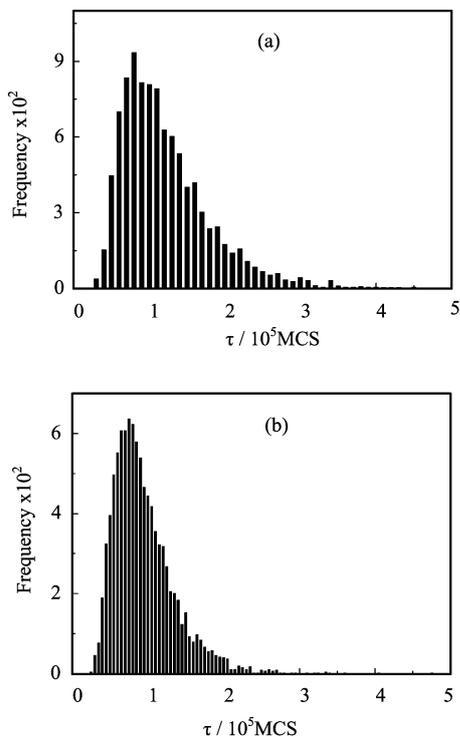


FIG. 1 Distribution of translocation time at (a) $u_0=0.6$ and (b) $u_0=1.0$ under the conditions of $N=40$, $a=1$, $\varepsilon_{AB}=0.1$, and $N_{trans}=0$.

In Fig.2 a crossover can be observed in the high-temperature region between the curves corresponding to $u_0=0.6$ and 1.0. In the lower-temperature region, the chains adopt compact conformations; E_s is very low and does not exert a significant effect on the translocation time. Under these conditions, the translocation time is governed mainly by the cohesive energy barrier E_c . Thus, when the absorbed energy is increased, the translocation time decreases. At high temperature, the chain exists in a coiled state with few connections between monomers. In this case, the cohesive energy barrier E_c is very low and does not significantly affect the translocation time. Therefore under these conditions, the translocation time is governed mainly by the entropy barrier E_s . When the strength of the absorbed energy is high, the movement of the polymer chain on the *trans* side, which tends to be confined within a two-dimensional space under such conditions, is restricted. As a result, the translocation time increases, which leads to the crossover between the two curves.

B. Variation of τ with respect to solvent quality

Considering scarcity of previous studies into the effects of solvent quality, the translocation time with respect to the solvent quality under the conditions $N=60$, $k_B T=1.0$, $a=1$, $N_{trans}=0$, and $u_0=1.0$ was studied, the results of which are shown in Fig.3. When the solvent

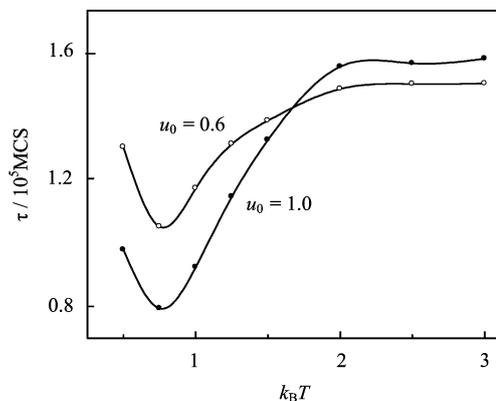


FIG. 2 Translocation time as a function of reduced temperature, under the conditions of $N=40$, $a=1$, $\varepsilon_{AB}=0.1$, and $N_{trans}=0$.

quality decreased from “good” to “poor” two distinct regions existed: for values of ε_{AB} lower than some critical interaction energy (ε_c) between the polymer and the solvent (where ε_c is the value of ε_{AB} corresponding to the minimum in τ), the value of τ decreases for increasing ε_{AB} ; when $\varepsilon_{AB} > \varepsilon_c$, the value of τ increases with increasing value of ε_{AB} . For $\varepsilon_{AB} < \varepsilon_c$, i.e., when the polymer chain is stretched, the value of E_c is extremely low and it does not affect the translocation time. In contrast, the value of E_s decreases upon increasing the value of ε_{AB} , causing the polymer chain to translocate through the pore more readily. Thus, the value of τ decreases. For $\varepsilon_{AB} > \varepsilon_c$, i.e., when the polymer chain is collapsed, the value of E_s affects the value of τ only slightly, so its influence can be neglected. In contrast, the value of E_c is a very important factor affecting the value of τ ; a pulling force must be applied to unwind the polymer by breaking the intermonomer interactions prior to its being dragged through the pore. When the solvent quality becomes poorer, the value of E_c increases. Under these conditions, the movement of the polymer chain translocating through the pore becomes more difficult and the translocation time is increased. This finding is unlike that in Section A because in that section a change in the amount of energy absorbed as a function of temperature was found, as is evident from Eq.(1), but in this case the absorbed energy was not affected by the quality of the solvent, i.e., the amount of energy absorbed in this section was constant.

C. Effect of the width of the interaction potential

After establishing the effect of the solvent quality on the translocation process, the influence that the width of the interaction potential has on the translocation time was determined. A series of simulations under the conditions $N=40$, $k_B T=1.0$, $\varepsilon_{AB}=0.1$, $N_{trans}=0$, and $u_0=0.6$ were performed. Figure 4 displays the obtained translocation time. It was observed that the

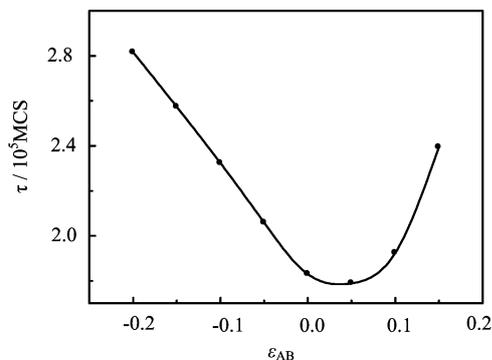


FIG. 3 Translocation time was plotted with respect to the solvent quality under the conditions of $N=60$, $k_B T=1.0$, $u_0=0.6$, and $N_{trans}=0$, $a=1$.

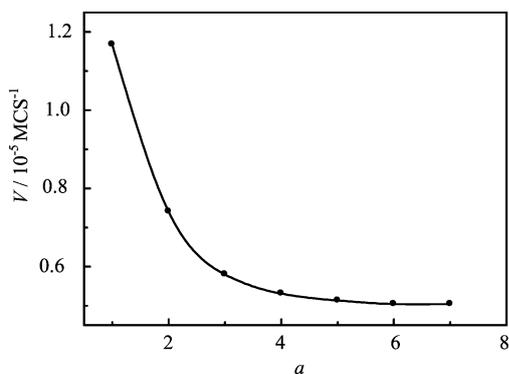


FIG. 4 Velocity with respect to the width of the interaction potential under the conditions of $N=40$, $u_0=0.6$, $\varepsilon_{AB}=0.1$, and $N_{trans}=0$ at a reduced temperature of $k_B T=1.0$.

translocation time decreased initially upon increasing the width of the interaction potential (a) before reaching a plateau. To explain this observation, the radius of gyration of the polymer chain was determined under these conditions: for $N=40$, a value for $\langle R_g^2 \rangle^{1/2}$ of 2.7 was obtained; i.e., $2\langle R_g^2 \rangle^{1/2} < 6$. When the width of the interaction potential is narrow, the movement of the polymer chain is restricted to two dimensions. Under these conditions the value of τ decreases upon increasing value of a . In contrast, when the width of the interaction potential is greater than double the root-mean-square radii of gyration of the chains, the polymer chain is not restricted. Therefore, the value of τ becomes stable. When the value of u_0 was changed, similar results were obtained.

D. Effect of the strength of the interaction potential

To determine the effect that the strength of the interaction potential between the monomer and the membrane on the *trans* side has on the translocation time, a series of simulations using different values of u_0 under the conditions $k_B T=1.0$, $N=40$, $a=1$, $N_{trans}=0$,

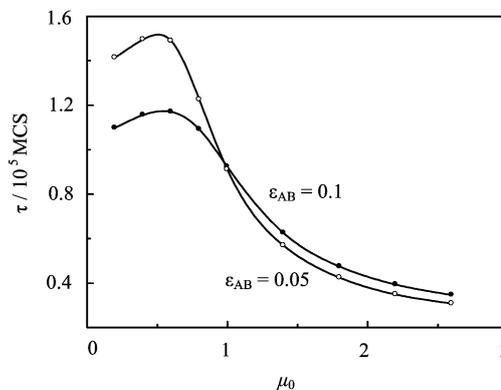


FIG. 5 Translocation time with respect to the strength of the interaction energy under the conditions of $N=40$, $k_B T=1.0$, $a=1$, and $N_{trans}=0$.

$\varepsilon_{AB}=0.1$, and $\varepsilon_{AB}=0.05$, were performed, as shown in Fig.5. Two distinct regions under the same set of solvent conditions were observed: when the strength of the absorbed energy u_0 is greater than some critical absorbed energy u_c (u_c is τ corresponding to maximum in this absorbed energy), the value of τ decreases with increasing absorbed energy u_0 . In contrast, when $u_0 < u_c$, the value of τ increases with increasing absorbed energy u_0 . These results suggest that when the amount of energy absorbed is high, the movement of the polymer chain is constrained in two dimensions on the *trans* side; at higher absorbed energy, the pulling force is stronger and, thus, the translocation time decreases. In contrast, for $u_0 < u_c$, the polymer chain tends to move in three dimensions, and under these conditions τ decreases. From Fig.5 it was observed that when the solvent quality was changed from “poor” to “comparatively good”, a crossover existed between values of ε_{AB} between 0.05 and 0.1. In contrast, when $u_0=0$, the translocation time was infinite. This result can be explained by considering Muthukumar’s nucleation model [11], which states that when a polymer chain has a certain number (above some critical value) of monomers on the *trans* side it will translocate fully through the pore. When $u_0=0$, it is very difficult to reach that critical number of monomer units on the *trans* side and, thus, the translocation time becomes infinite.

E. Dependence of the translocation on polymer length

To determine the effect of the value of N on the translocation time and velocity in the “poor” solvent, a series of simulations on polymers of different lengths at reduced temperature ($k_B T=1.0$) under the conditions $a=1$, $\varepsilon_{AB}=0.1$, and $N_{trans}=0$ were performed, where the thickness of the pore was $W=1$. These calculations were processed at values of u_0 of 2.0 and 0.4, respectively. Figure 6 displays the results. In this simulation by using this model, an adsorption threshold at

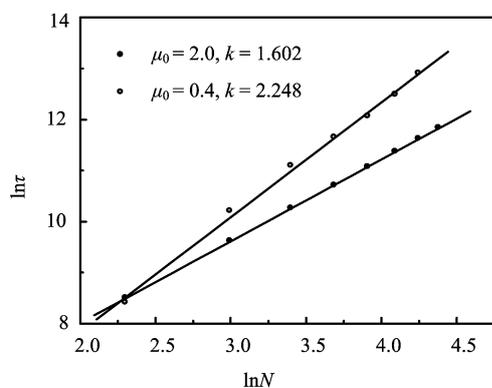


FIG. 6 Log-log plot of the translocation time (τ) as a function of the chain length (N), under the conditions $k_B T=1.0$, $a=1$, $W=1$, $N_{trans}=0$, and $\varepsilon_{AB}=0.1$.

$u_c \approx 0.62$ for the “mushroom”-to-“pancake” adsorption transition was obtained [34]. When u_0 was 2.0, i.e., above the value of u_c , the slope of the line was 1.602; when u_0 was 0.4, i.e., below u_c , the slope of the line was 2.248. This result is consistent with the conclusions drawn by Chuang, Kantor, and Kardar [32] and Binder [20], but it is at variance with those of Muthukumar [10] ($\tau \propto N^2$) and Park and Sung [19] ($\tau \propto N^3$). Clearly, values of $\tau \propto N^3$ would imply that a single pore in a thin membrane is as severe an obstacle to polymer motion as many entanglements in a dense polymer melt, which seems implausible; on the other hand, values of $\tau \propto N^2$ would imply that the translocating chain relaxes faster (in the presence of an excluded volume) than an unconstrained chain in dilute solution, which also is implausible [20]. Figure 7 displays the average translocation velocity with respect to the polymer chain length under the conditions $W=1$, $a=1$, $k_B T=1.0$, $\varepsilon_{AB}=0.1$, $N_{trans}=0$, and $u_0=2.0$. When N was less than 100, the velocity decreased upon increasing the value of N . For $N \geq 100$, the velocity reached a plateau, i.e., it was independent of the value of N . This result is in agreement with the findings reported by Meller *et al.* [4].

F. Variation of translocation probability with N_{trans}

Both the translocation time and the translocation probability are very important factors in determining the translocation efficiency. The definition of the translocation probability is the number of samples that have translocated successfully through the pore divided by the total number of samples. A series of simulations using a fixed polymer length ($N=40$), a temperature $T=1.0$, and conditions of $u_0=0.6$, $W=1$, and $a=1$ at values of $\varepsilon_{AB}=0.2, 0.1, -0.1$, and -0.2 were performed. Figure 8 displays the obtained probabilities. The resulting translocation probability increased upon increasing the value of N_{trans} until it reached a maximum. This finding is in agreement with the classical nucleation the-

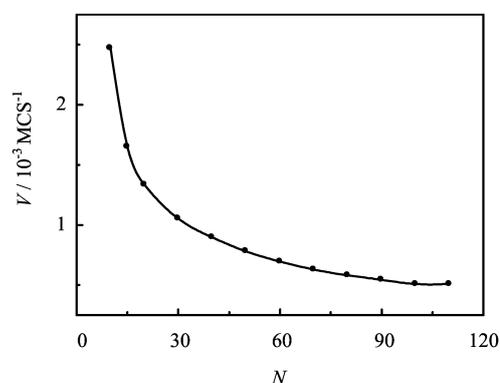


FIG. 7 The velocity (V) with respect to the number of monomers (N), under the conditions $W=1$, $a=1$, $\varepsilon_{AB}=0.1$, $N_{trans}=0$, $k_B T=1.0$, and $u_0=2.0$.

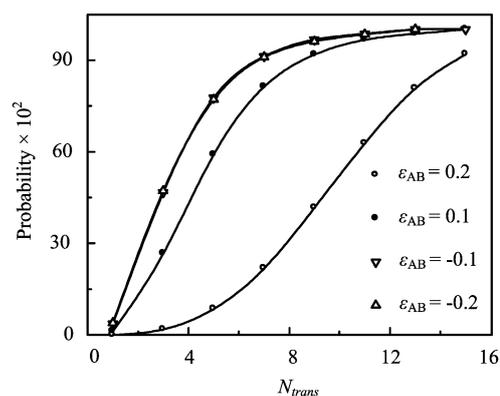


FIG. 8 The translocation probability with respect to N_{trans} , for a polymer with chain length $N=40$ under conditions of $a=1$, $W=1$, and $u_0=0.6$, and at a reduced temperature of $k_B T=1.0$.

ory presented by Muthukumar [11]. When the solvent quality improved from “poor” to “good” the translocation probability reached its maximum more rapidly. This phenomenon occurred because in a “poor” solvent, the polymer chain exists in a collapsed state. For such a polymer chain to translocate through the pore it must first unwind, but such a conformation is unstable. Thus, it prefers to maintain its collapsed state, which results in a low translocation probability. In a “good” solvent, however, the polymer chain exists in a stable unwound state. Therefore the translocation probability is higher than that of the polymer in the “poor” solvent.

IV. CONCLUSION

A minimalist model was proposed for polymers which can be used to simulate the translocation of a flexible polymer through a pore in a membrane, induced by its adsorption on the *trans* side of the membrane. When the temperature varied, it was found that the transition occurred. At $T < T_c$, τ decreased upon in-

creasing the temperature. At $T > T_c$, τ increased upon increasing the temperature. This result was explained by considering the effects of the values of E_c and E_s . When the width of the interaction potential varied, it was found that the translocation time of the polymer chain decreased initially upon increasing the width of the field. However when the width of field (a) was above $2\langle R_g^2 \rangle^{1/2}$, the translocation time became stable because the polymer chains that translocated through the pore could move freely in three dimensions on the *trans* side. The variation of τ with respect to the quality of the solvent was also studied. When the solvent quality worsened from “good” to “poor”, there was a transition point. For $\varepsilon_{AB} < \varepsilon_c$, the value of τ decreased upon increasing the value of ε_{AB} , whereas for $\varepsilon_{AB} > \varepsilon_c$, τ increased with increasing ε_{AB} . The values of E_c and E_s were also considered in explaining these phenomena. In addition, the scaling behavior of the translocation time τ with respect to chain length was evaluated. Below the adsorption threshold ($u_0 < u_c \approx 0.62$ in our model) it was found that τ was proportional to $N^{2.248}$. Above the adsorption threshold, values of $\tau \propto N^{1.602}$ was obtained. An interesting result relating to the velocity of the translocating chain was obtained: when the length of the translocating chain was not too long, the value of V decreased with increasing values of N . Otherwise, V reached a plateau, i.e., it was independent of the value of N . These results are in agreement with the findings reported by Meller *et al.* [4]. Finally, the translocation probability with respect to the value of N_{trans} and the solvent quality was studied. It was found that when N_{trans} was constant, the probability increased initially upon improving the solvent quality from “poor” to “good” before it reached a stable value.

V. ACKNOWLEDGMENTS

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