

Surface-adsorption-induced polymer translocation through a nanopore: Effects of the adsorption strength and the surface corrugation

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The surface corrugation plays an important role in single polymer diffusion on attractive surfaces. However, its effect on dynamics of surface adsorption-induced polymer translocation through a nanopore is not clear. Using three-dimensional Langevin dynamics simulations, we investigate the dynamics of a flexible polymer chain translocation through a nanopore induced by the selective adsorption of translocated segments onto the *trans* side of the membrane. The translocation probability P_{trans} increases monotonically, while the mean translocation time τ has a minimum as a function of the adsorption strength ε , which are explained from the perspective of the effective driving force for the translocation. With the surface being smoother, τ as well as the scaling exponent α of τ with the chain length N decreases. Finally, we show that the distributions of the translocation time are non-Gaussian even for strong adsorption at a moderate surface corrugation. A nearly Gaussian distribution of the translocation time is observed only for the smoothest surface we studied.

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I. INTRODUCTION

The transport of polymers through a nanopore embedded in membranes has attracted considerable attention in experiments [1–12], analytical theories and simulations [13–40]. Polymer translocation is a typical nonequilibrium process, and is also related to many crucial processes in biology, such as the passage of mRNA through nuclear pores, the translocation of proteins through the endoplasmic reticulum, and the viral injection of DNA into a host cell. In addition, the translocation processes have potential revolutionary technological applications, such as rapid DNA or RNA sequencing [1,6,7,10], gene therapy [41], and controlled drug delivery [42].

Due to the loss of a great number of available chain conformations, polymer translocation faces a large entropic barrier and thus the driving forces are needed. The driving mechanism for translocation has been a subject of extensive discussions. One main resource of the driving force both *in vivo* and *in vitro* is the transmembrane electrical potential. In biological cells, it comes from the transmembrane electrical potential since biopolymers, such as single-stranded DNA (ssDNA), are negatively charged. A chemical potential gradient across the membrane can also provide a driving force. Examples include different solvent conditions on the two sides of the membrane [26–28], the binding particles (BPs) existing on the *trans* side [29,30], and the selective adsorption of chains on the *trans* side of the membrane [31–33]. In addition, polymer translocation can be driven by the geometrical confinement of the chain [34]. Theoretically, the tension propagation theory was proposed and developed to depict the nonequilibrium aspects for the case of driven translocation [35–37].

As to the translocation induced by the selective adsorption, Milchev *et al.* [32] have suggested that the scaling behavior of the mean translocation time with the chain length depends on the adsorption strength ε . Recently, Yang and

Neimark [33] have considered the translocation driven by polymer adsorption to the inner surface of a confining wall by using the self-consistent field theory. In both studies, the adsorption strength ε enters as an important parameter that could mediate the translocation dynamics. Obviously, the adsorption-induced polymer translocation should strongly depend on the diffusion dynamics of an adsorbed chain on the surface. However, the surfaces are assumed to be perfectly flat in above studies, which is not realistic. Particularly, Mukherji *et al.* [43] have reported that the atomic-scale corrugation breaks in-plane translational Galilean invariance as finite free-energy corrugation barriers prevent the chains from free lateral sliding. As a consequence, the in-plane center-of-mass diffusion of adsorbed chains depends on the extent of the surface corrugation [43]. Furthermore, a more recent study by Carr *et al.* [44] has shown that a change in the atomic scale surface roughness and its charge density could alter the Langmuir constant of a small organic molecule by three orders of magnitude. It implied that subtle variations in the surface corrugation could give rise to variations of the adsorption strength along the surface. Thus, the surface corrugation should have an effect on the dynamics of polymer translocation induced by surface adsorption, particularly in the case of strong adsorption where the translocated chains show pancake-like conformations.

To this end, we investigate how the surface parameters, including the corrugation and the adsorption strength, affect the polymer translocation in this work. The outline of this paper is as follows. We briefly describe our model and the simulation technique in Sec. II. Our results and corresponding discussions are presented in Sec. III. Finally, we give a summary in Sec. IV.

II. MODEL AND METHODS

The three-dimensional model geometry we consider in this work is represented in Fig. 1(a), where a coarse-grained polymer chain is translocating through a nanopore induced by the selective adsorption of the chain on the *trans* side of

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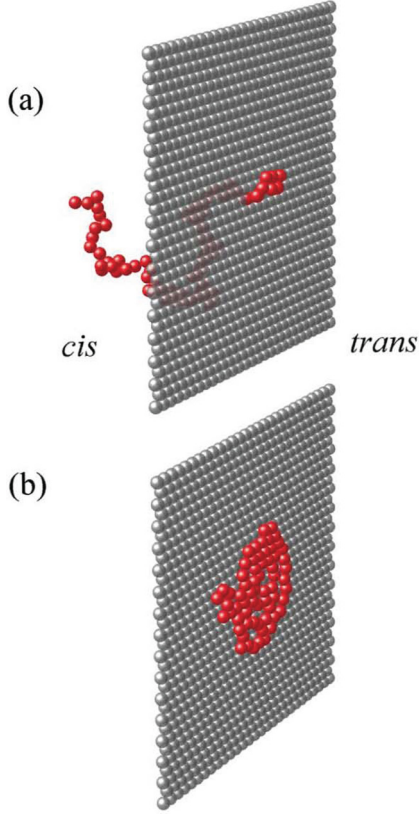


FIG. 1. (Color online) Schematic representation of a polymer chain of length $N = 64$ (a) translocation through a nanopore induced by the selective adsorption of chain segments on the *trans* side of the membrane (b) adsorbed on the *trans* side of the membrane just after the translocation.

the membrane. As illustrated in Fig. 1(b), the chain shows a pancake-like conformation on the membrane just after the translocation.

In our simulations, the polymer chain is modeled as a bead-spring chain of Lennard-Jones (LJ) particles, and each bead represents a segment. Each pair of nonbonded particles is subjected to the short-range repulsive LJ potential:

$$U_{\text{LJ}}(r) = \begin{cases} 4\varepsilon_0[(\sigma/r)^{12} - (\sigma/r)^6] + \varepsilon_0, & r \leq r_{\text{cutoff}} \\ 0, & r > r_{\text{cutoff}}. \end{cases} \quad (1)$$

Here, σ is the diameter of a segment. ε_0 is the depth of the potential, and $r_{\text{cutoff}} = 2^{1/6}\sigma$ is the cutoff distance. The connectivity between neighboring segments is achieved by applying the finite extension nonlinear elastic (FENE) potential,

$$U_{\text{FENE}}(r) = -\frac{1}{2}kR_0^2 \ln(1 - r^2/R_0^2), \quad (2)$$

where r is the distance between consecutive segments, k is the spring constant, and R_0 is the maximum allowed separation between connected segments.

The membrane consists of a (111) surface of a face-centered-cubic solid. Immobile particles in the membrane are static with a distance of 1.209σ [43]. The nanopore embedded in the membrane is orthohexagonal with a side length 1.209σ . At the *cis* side, the surface is repulsive for a chain. The

interaction between the segment and the membrane surface is also described by the above short-range repulsive LJ potential with the interaction strength ε_0 , the diameter of LJ particle $\sigma_{\text{sp}} = \frac{\sigma + \sigma_w}{2}$, where σ_w is the diameter of the surface particles, and $r_{\text{cutoff}} = 2^{1/6}\sigma_{\text{sp}}$. Meanwhile, the *trans* side of the surface is attractive for chain segments. In this situation, the interaction between the segment and the membrane surface is described by a long-range attractive LJ potential with the interaction strength ε , the diameter of LJ particle σ_{sp} , and $r_{\text{cutoff}} = 2.5\sigma_{\text{sp}}$. By changing the diameter of the surface particles σ_w , σ_{sp} is an adjustable parameter which mediates the surface corrugation and ranges from 0.95 to 1.15σ in this work. Larger values of σ_{sp} result in smaller surface corrugation. The range of ε is 1.5 – $3.3\varepsilon_0$ to characterize different adsorption strengths.

The motion of chain segments is described by the Langevin equation:

$$m\ddot{\mathbf{r}}_i = -\nabla U_i - \xi\mathbf{v}_i + \mathbf{F}_i^R. \quad (3)$$

Here, m , ξ , and \mathbf{v}_i are the bead's mass, friction coefficient, and velocity, respectively. $-\nabla U_i$ and $-\xi\mathbf{v}_i$ are the conservative and frictional forces exerted on the segments, respectively. \mathbf{F}_i^R is the random force that satisfies the fluctuation-dissipation theorem [45].

In our simulations, the LJ parameters ε_0 , σ , and the bead mass m determine the system energy, length, and mass units, leading to the corresponding time scale $t_{\text{LJ}} = (m\sigma^2/\varepsilon_0)^{1/2}$ and force scale ε_0/σ , which are of the order of ps and pN, respectively. The dimensionless parameters in this work are then chosen to be $R_0 = 2$, $k = 7$, $\xi = 0.7$. We set $k_B T = 1.2\varepsilon_0$ with $k_B T$ being the thermal energy. Then the Langevin equation is integrated in time by the method proposed by Ermak and Buckholz [46].

Initially, for the chains of length $N = 32$ – 128 , the first seven segments are placed at the *trans* side of the membrane and kept adsorbed on it to improve the simulation efficiency. During the relaxation process, the eighth segment is fixed at the center of the pore with the remaining chain segments being under thermal collisions described by the Langevin thermostat. As the chain relaxation completes, the eighth segment is released and the translocation event begins, which is considered to be successful when the last monomer exits the pore. Typically, each data point is obtained by averaging over 1000 successful translocation events to reduce statistical errors.

III. RESULTS AND DISCUSSIONS

A. Effect of the adsorption strength on the translocation dynamics

After one end of a chain is captured by the nanopore, the chain needs to overcome the energy barrier for translocation due to the loss of conformational entropy. In the present work, the translocation of a chain is induced by the selective adsorption of translocated segments on the *trans* side of the membrane, which decreases the free energy of the chain. The energy gain of a segment in contact with the surface provides a driving force for the translocation. Therefore, the adsorption strength ε is a crucial factor affecting the translocation dynamics. Next, we study this effect by measuring the translocation

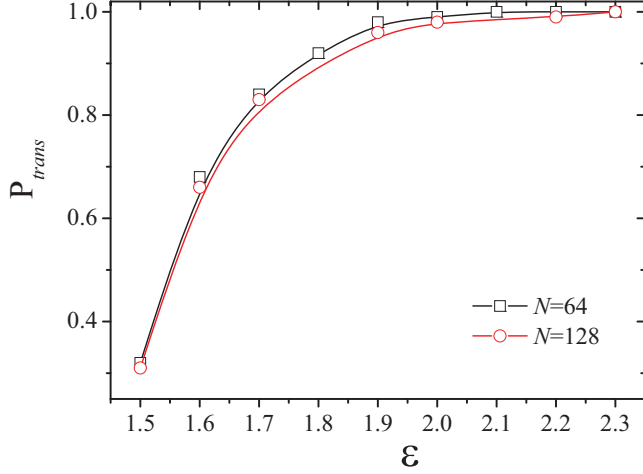


FIG. 2. (Color online) The translocation probability P_{trans} as a function of the adsorption strength ε for two different chain lengths $N = 64$ and 128.

probability P_{trans} and the mean translocation time τ at a fixed corrugation $\sigma_{sp} = 1$.

The translocation probability P_{trans} is defined as the ratio of the successful translocation events to all attempts in the simulations. Figure 2 shows that P_{trans} increases rapidly first and then saturates gradually with increasing ε . This can be well understood from the perspective of the effective driving force F_{eff} written as

$$F_{eff} = F_{ad}(\varepsilon) - F_{cis,en}(s) - F_{trans,en}(s,\varepsilon). \quad (4)$$

Here, $F_{ad}(\varepsilon)$ is the force from adsorption, which increases with ε . $F_{cis,en}(s)$ and $F_{trans,en}(s,\varepsilon)$ is the entropic force from the *cis* and *trans* sides with s being the translocation coordinate, respectively. With increasing ε , $F_{ad}(\varepsilon)$ dominates the translocation process, leading to the observed behavior of P_{trans} .

However, is the increase of ε always beneficial to the translocation process? An obvious fact is that the diffusivity of translocated segments relies on ε . When the adsorption is too strong, these segments cannot diffuse away from the pore in time. This crowding effect leads to a resistance force hindering the translocation $F_{trans,en}(s,\varepsilon)$, as reflected by Eq. (4). With the adsorption being stronger, $F_{trans,en}(s,\varepsilon)$ should get larger. With increasing ε , the interplay between $F_{ad}(\varepsilon)$ and $F_{trans,en}(s,\varepsilon)$ leads to a nonmonotonic dependence of F_{eff} on ε . As shown in Fig. 3, the mean translocation time τ does have a minimum as a function of ε for the studied two chain lengths. Similar behaviors of τ with varying ε were observed by Yang and Neimark [33].

The chain length dependence of τ ($\tau \sim N^\alpha$) for different ε is plotted in Fig. 4. We observe that α equals to 1.62 ± 0.02 for $\varepsilon = 1.6$ and 2.2, and crosses over to 2.53 ± 0.03 for $\varepsilon = 3.3$. The scaling exponent $\alpha = 1.62 \pm 0.02$ for $\varepsilon = 1.6$ and 2.2 is in agreement with the scaling prediction $\tau \sim N^{1+2\nu-\varphi}$ by Milchev *et al.* [32] within the statistical errors, where $\nu = 0.588$ is the Flory exponent in three dimensions (3D), and $\varphi = 0.50$ is the crossover exponent [32]. In addition, this scaling exponent is also close to $1 + \nu$ for driven translocation under a chemical potential difference across

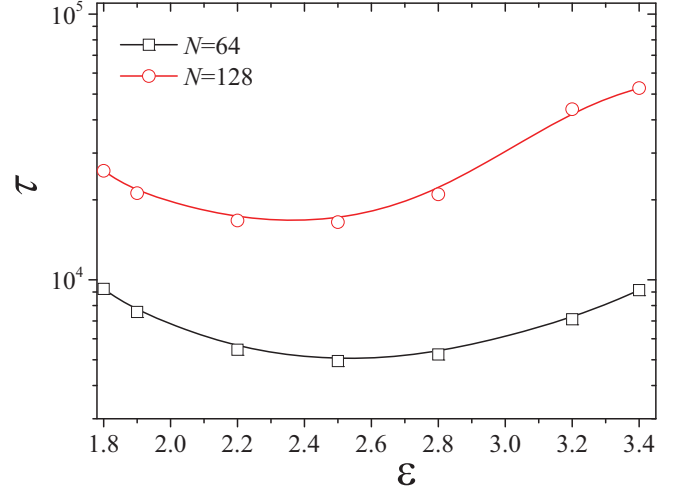


FIG. 3. (Color online) The mean translocation time τ as a function of the adsorption strength ε for two different chain lengths $N = 64$ and 128.

the membrane [20]. As ε increases to 3.3, the translocated segments are strongly adsorbed on the membrane and thus the translocation dynamics is dominated by the in-plane chain diffusion. The corresponding scaling relation $\tau \sim N^{2.53 \pm 0.03}$ is consistent with the scaling behavior of the Rouse relaxation time for a chain in two dimensions (2D), $\tau_R \sim N^{1+2\nu_{2D}} = N^{2.50}$, with $\nu_{2D} = 0.75$ being the Flory exponent in 2D.

B. Dependence of the translocation dynamics on the surface corrugation

Besides the adsorption strength ε , it has been suggested in the previous work by Mukherji *et al.* [43] that the diffusivity of a strongly adsorbed chain is significantly affected by the surface corrugation. Therefore, the surface corrugation should play an important role in the translocation dynamics of a chain, which has not yet been studied. In the following, the surface corrugation is tuned by varying the LJ radius σ_{sp} used

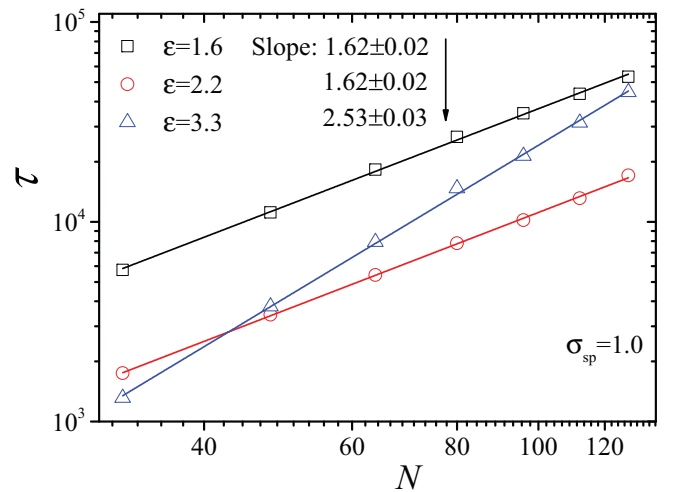


FIG. 4. (Color online) The chain length dependence of the mean translocation time τ , $\tau \sim N^\alpha$, at different adsorption strengths.

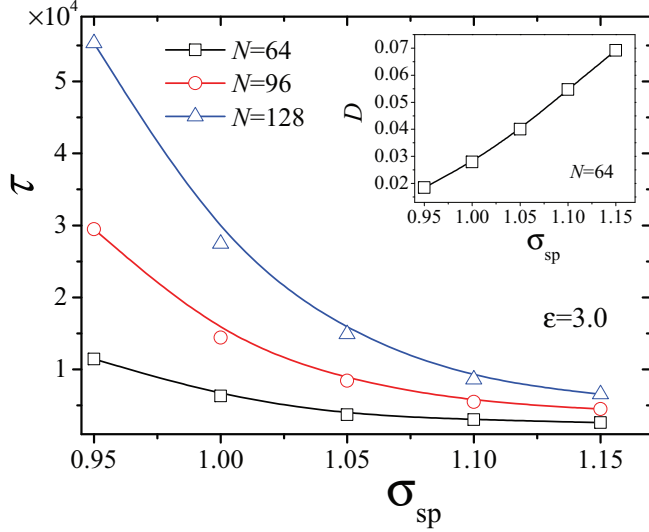


FIG. 5. (Color online) Influence of the surface corrugation σ_{sp} on the mean translocation time τ for chains of different lengths. The inset shows the dependence of the chain surface diffusion coefficient D on σ_{sp} . Here, the adsorption strength is kept at $\varepsilon = 3.0$.

in the potential between segments and surface particles from 0.95 to 1.15, as has been done previously [43]. Note that the corrugation is reduced with increasing σ_{sp} . The adsorption strength is kept at $\varepsilon = 3.0$ unless otherwise stated.

Figure 5 shows that τ monotonically decreases with decreasing surface corrugation for chains of different lengths. In order to understand this behavior, we calculate the in-plane diffusion coefficient D of a chain strongly adsorbed onto the surface by measuring the slope of the mean-square displacement (MSD) of its center of mass at a long time limit. As shown in the inset of Fig. 5, D increases with increasing σ_{sp} , indicating that a smoother surface is in favor of the in-plane chain diffusion. This phenomenon was also observed by Mukherji *et al.* [43]. As mentioned above, for strong adsorption, the surface diffusion of a chain is of essential importance to the translocation dynamics. It is the faster surface diffusion of the chain that accelerates the translocation.

Figure 6 shows the effect of the surface corrugation on the scaling behaviors of $\tau \sim N^\alpha$ with the extracted scaling exponent α being plotted in the inset. As σ_{sp} increases from 0.95 to 1.15, α decreases from 2.39 ± 0.06 to 1.34 ± 0.01 . Longer chain lengths give rise to smaller in-plane diffusion coefficient D , and thus the increase of D due to the smoother surface appears to be relatively favorable for the translocation of longer chains. As shown in Fig. 5, we observe a faster decay rate of τ with increasing σ_{sp} for longer chains. These different decay rates explain the decrease of α as a function of σ_{sp} .

C. Translocation time distribution

Finally, we turn to the distributions of the translocation time normalized by their average value t/τ . As plotted in Fig. 7(a), compared to the translocation driven by a strong transmembrane electric field [20,38,39] or a large pulling force exerted on the first segment [40], we find that even for the cases of strong adsorption, the distributions are still non-Gaussian,

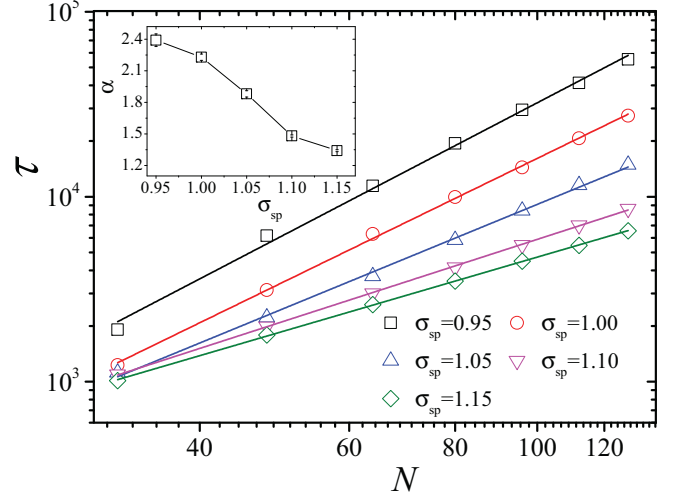


FIG. 6. (Color online) The scaling behaviors of $\tau \sim N^\alpha$ for different surface corrugations σ_{sp} . The extracted scaling exponent α as a function of σ_{sp} is plotted in the inset. Here, the adsorption strength is kept at $\varepsilon = 3.0$.

reflecting that the translocation induced by strong adsorption is a weakly driven process due to the crowding effect near the pore. The standard deviation (SD) of the distribution has a minimum $SD_{t/\tau} = 0.246$ at $\varepsilon = 2.5$ when the translocation is the fastest.

Figure 7(b) shows that the distribution gets narrower with $SD_{t/\tau}$ decreasing monotonically as the surface becomes smoother. Moreover, a nearly Gaussian distribution of the translocation time is observed at $\sigma_{sp} = 1.15$. In this case, the surface diffusion of the chain is fast enough so that the crowding effect near the pore is of marginal importance for the translocation.

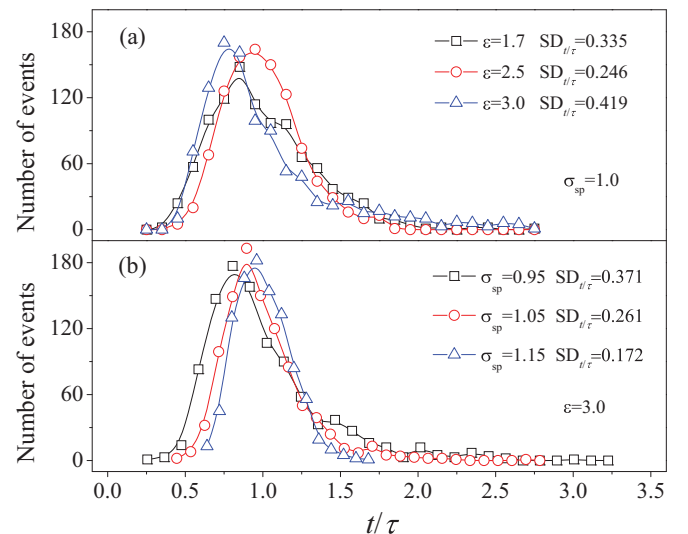


FIG. 7. (Color online) Distributions of the translocation time normalized by their average value t/τ under (a) different adsorption strengths ε and (b) different surface corrugations σ_{sp} . Here, the chain length is $N = 64$.

IV. CONCLUSIONS

To summarize, we have investigated the dynamics of a flexible polymer chain translocation through a nanopore induced by the selective adsorption of translocated segments onto the *trans* side of the membrane by using 3D Langevin dynamics simulations. We find that the translocation probability P_{trans} increases monotonically, while the mean translocation time τ has a minimum as a function of the adsorption strength ε . These effects of ε on the translocation dynamics are understood from the perspective of the effective driving force for the translocation. As to the effect of the surface corrugation on the translocation dynamics, we show that τ as well as the scaling exponent α of $\tau \sim N^\alpha$ decreases with the surface being smoother. Finally, the distributions of the translocation time are non-Gaussian even for the case of strong adsorption at a moderate surface corrugation $\sigma_{sp} = 1.0$. For the smoothest surface we studied, a nearly Gaussian distribution of the translocation time is observed.

We have noticed several experimental works about the effects of surface properties on the translocation process of a polymer chain. A recent work by Shankla and Aksimentiev

[11] has shown that the electrically biased graphene membranes exhibit exquisite control over the adsorption of ssDNA. This property has an obvious impact on the conformations of the translocating ssDNA, which can be used to realize stop-and-go nanopore translocation of ssDNA [11]. Furthermore, Banerjee *et al.* [12] have reported that by using multiple stacked graphene layers, the translocation of ssDNA is slowed down due to the variations in the interaction between ssDNA and the membrane. Therefore, it is possible to realize the translocation of a polymer chain induced by asymmetric surface adsorption in experiments through adopting proper membrane structures and/or varying the surface properties of the membranes.

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