Dynamics of a Polymer in the Presence of Permeable Membranes

Hyoungsoo Yoon    J.M. Deutsch
University of California, Santa Cruz
Santa Cruz, CA 95064
October 16, 2018

Abstract

We study the diffusion of a linear polymer in the presence of permeable membranes without excluded volume interactions, using scaling theory and Monte Carlo simulations. We find that the average time it takes for a chain with polymerization index \( N \) to cross a single isolated membrane varies with \( N \) as \( N^{5/2} \), giving its permeability proportional to \( N^2 \). When the membranes are stacked with uniform spacing \( d \) in the unit of the monomer size, the dynamics of a polymer is shown to have three different regimes. In the limit of small \( d \ll N^{1/2} \), the chain diffuses through reptation and \( D \sim N^{-2} \). When \( d \) is comparable to \( N^{1/2} \) the diffusion coefficients parallel and perpendicular to the membranes become different from each other. While the diffusion becomes Rouse-like, i.e. \( D \sim N^{-1} \), in the parallel direction, the motion in the perpendicular direction is still hindered by the two-dimensional networks. The diffusion eventually becomes isotropic and Rouse-like for large \( d \gg N \).

1 Introduction

The physics of tethered membranes\cite{1,2,3,4} has been studied with great interest due not only to its relevance to biology but also to its numerous applications in technology. Stabilizing liposomes, which are used in drug delivery, by tethered membranes is just one of many important applications\cite{5}. It is also possible that tethered membranes could provide a new
way of filtrating polymers in a solution according to their molecular weights. This method should be especially useful where conventional methods such as gel electrophoresis[6, 7, 8] do not work.

Motivated by this, we study in this paper the effects of permeable membranes on the dynamics of a flexible linear polymer. More specifically we focus on the diffusion characteristics of a linear polymer in the presence of a single or multiple membranes.

We model our permeable membranes as two-dimensional regular networks of polymers with the mesh size bigger than the persistence length of the diffusing chain. This type of membranes can be made by crosslinking linear polymers using ultraviolet light[5] or by extracting spectrin networks from human red blood cells[9]. The widely used molecular filters, called zeolites[10], may also provide a good system if the molecular parameters such as the Kuhn length of the diffusing polymer is sufficiently small.

For simplicity, we restrict our discussion to systems with no excluded volume interactions. The only constraints the two-dimensional networks impose on the dynamics of the chain polymer are topological barriers. From a theoretical point of view, this is a new problem quite distinct from the study of reptating chains in an inhomogeneous environment[11, 12], which has been investigated extensively partly due to its relevance to DNA gel electrophoresis. Even though the main ingredients in reptation theory are the topological constraints imposed by surrounding chains or gel networks, most studies on reptation in an inhomogeneous medium concentrates on more complex (and presumably more realistic) systems with excluded volume interactions.

The remainder of this paper is organized as follows. In section 2 we first define our problem in more precise terms and give a brief account of the simulation methods used in this work. In section 3 we consider the longest relaxation time of a linear polymer in the presence of a single membrane. This is obtained numerically by measuring the first passage times of the linear polymer through an array of membranes which are placed with a spacing proportional to the radius of gyration of the polymer. In section 4, we use the data obtained in the previous section to calculate the permeability for our two-dimensional network, which is one of the most important characteristics of permeable membranes. In section 5, we study
the multiple membrane case with uniform spacings in more detail. The diffusion constants parallel to the membranes as well as the perpendicular ones are studied. We first present a simple scaling theory as a function of the polymerization index and the spacing between the membranes. Then we present the simulation results which support this analysis. In section 6, we consider the mean square displacements of a monomer as a function of time when a chain disengages from a network. Finally in section 7 we summarize our results and discuss possible applications of this study.

2 The Model

We consider a random walk chain of \( N \) monomers or beads on a cubic lattice with a lattice spacing \( b \), which is the monomer size or the bond length of the chain. We only consider even \( N \)’s due to the bipartite nature of the cubic lattice.

The simulation is done by allowing the chain to move according to the following set of Monte Carlo rules which mimics Rouse dynamics.

I Two consecutive bonds which form an \( L \)-shape can be inverted, in the plane formed by the \( L \).

II A kink, two consecutive bonds which are on top of each other, can be flipped to one of the six lattice directions including the original one. (See, for example, Fig. 1 of reference [13].) The free ends of the chain are treated as kinks.

Now we consider another cubic lattice with lattice spacing \( a \) which interpenetrates the first one. We assume \( a \) is an integral multiple of \( b \) so that the lattice points of the second lattice always reside at the centers of the unit cells of the first. We let \( a = b = 1 \) in the numerical simulations.

Our membrane is a two-dimensional slice of the second lattice in the horizontal plane. Since the membrane occupies a different lattice than that of the chain, it does not alter the statics of the chain. But we assume that the chain and the membranes cannot pass through each other. That is, the links of the membranes act as topological barriers for the motion of the chain.
This is implemented in our simulation by modifying rule I. We allow the $L$-inversion only when the chain does not pass through a link of a membrane by this move. This is basically the same as the Doi-Evans-Edwards algorithm\cite{14, 15}. Note that the dynamics of a chain is altered only near the vicinity of the membranes and if the plane formed by the $L$ is horizontal rule I is not affected at all because the links of the membranes are all horizontally placed.

3 Relaxation time of chain penetration

First we study the effects of a single membrane on the dynamics of a linear polymer with polymerization index $N$. One of the most basic questions we have to ask in this situation is how much the membrane will slow down the diffusion of the polymer which is in its vicinity. More specifically we ask what is the time scale of the complete penetration of the polymer through the two-dimensional network.

Before we define the relaxation time for this penetration, we point out some problems associated with the very concept of the “complete penetration”. First we note that because a polymer has a finite size of order the radius of gyration, $R_g \sim N^{1/2}b$, the presence of the membrane interferes with the Rouse dynamics of the chain as long as its center of mass lies in a slab-like region around the membrane. We call this region of thickness $\sim R_g$ the entanglement zone. Here we stress that the thickness of the entanglement zone is a function of $N$.

If we try to define the relaxation time of a chain through a membrane by diffusion, we soon encounter difficulties. Diffusion is a random process and the polymer may never pass through the membrane. If the polymer penetrates the membrane at some point in time, the question remains as to how much time should be attributed to the effect of the membrane.

We circumvent this problem in the following way. First we consider two configurations, one with the center of mass of the polymer at a distance $\sim R_g$ from the membrane and another in which the polymer is maximally entangled (on average) with the membrane, that is, its center of mass lies at the membrane. Since the equilibrium statics are not affected by the topological membrane, these two configurations should arise with the same probability in equilibrium. Then from the principle of detailed balance, the transition probability from
one configuration to the other should be the same as the reverse. Hence the transition times in both directions are the same.

If we place a chain in the maximally entangled position (as defined in the previous paragraph) the chain eventually diffuses out to either end of the entanglement zone. We define the average first passage time in this process as the escape time. Then we define the penetration time as (4 times) the escape time since, as described above, the escaping process takes the same amount time as the reverse process and the penetration is, by definition, the combination of the two. Note that the chain does not necessarily penetrate to the other side of the membrane in this time scale.

The penetration time defined this way can be estimated easily by simple application of the reptation theory\[16\]. First we note that the chain typically crosses the membrane \(\sim N^{1/2}\) times in the entanglement zone with the average contour length \(\sim N^{1/2}b\) between the crossing points, or the entanglement points. The relaxation of the chain can be divided into two processes; Rouse relaxation of chain segments between entanglement points, which we call Rouse segments, and diffusion of kinks past entanglement points.

Even though this situation is quite different from the one of the usual reptation theory described by “reptilian” motion in a tube, we can still imagine the chain wiggling through a series of entanglement points inside an imaginary tube, its mean diameter being of order \(N^{1/4}b\).

The longest relaxation time in reptation theory is \(\tau_1 N^3/N_e\), where \(N_e\) is the number of monomers between entanglement points. Since \(N_e \sim N^{1/2}\) in our case, we obtain\[16\]

\[
\tau_m \sim N^{5/2}\tau_1
\]

Therefore dynamics of a chain through a tethered membrane is faster than the reptation in a three-dimensional network \(\tau_{\text{rep}} \sim N^3\tau_1\) and slower than the Rouse diffusion \(\tau_R \sim N^2\tau_1\), as expected. Note that \(N^{5/2}\)-dependence has been known to exit in various contexts such as the dynamics of a ring polymer in a gel\[17\].

In a time scale of \(\tau_m\) the chain moves a distance of the order \(R_g \sim N^{1/2}b\). Hence if we place an infinite array of membranes with \(\sim R_g\) apart from one another then the average time for a chain to hop across a membrane should be proportional to the relaxation time \(\tau_m\).
We conducted a simulation under these conditions for chain lengths 8 through 80. The inter-
membrane distances are set to $2bN^{1/2}$, with $b = 1$. At these distances the chains touching
two adjacent membranes at the same time is highly unlikely. The penetration time, $\tau \sim \tau_m$
is numerically defined as follows.

\[
\frac{1}{\tau} = \left\langle \frac{1}{\tau_p} \right\rangle
\]

where $\tau_p$ is the first passage time from the center of one inter-membrane zone to a neighboring
one. The angular brackets indicate the average over the distribution of $\tau_p$’s.

The log-log plot of $\tau$ vs. $N$ is shown in Fig. 1. The typical number of $\tau_p$’s used in obtaining
each point is approximately $10^4$. As the figure shows the exponent is clearly close to 2.5 as
the above scaling theory predicts. If there were no fluctuations in the size of the chain, we
should be able to produce more accurate results by shrinking the intermembrane distance
down to the radius of gyration of the chain. Because of the more generous spacing used in
the simulation the results must contain a part from Rouse dynamics. But the effect should
be small because $\tau_m \gg \tau_R$ for large $N$.

The penetration time is also expected to depend on the mesh size of the membrane, $a$.
Even though the $a$ dependence is harder to ascertain in our lattice models since $a$ only takes
integer values, it can be obtained using simple scaling argument as follows. First we assume
$\tau_m$ is a function of only $N$ and $a/b$. And since we already know the $N$-dependence we write
$\tau_m$ as

\[
\tau_m = \tau_1 N^{5/2} \left( \frac{a}{b} \right)^p
\]

where $p$ is the scaling exponent. When $a$ approaches $N^{1/2}$ the dynamics should be Rouse-
like. Hence we get $p = -1$. That is, $\tau_m$ is linearly proportional to the inverse of the mesh
size $a$ unlike $\tau_{rep}$, which has the quadratic dependence on $a^{-1}$.

4 Permeability of a membrane

As suggested earlier, two-dimensional networks of polymers or two-dimensional slabs with
pores could be used for filtration purposes. One of the most fundamental quantities in the
membrane filtration technique is the permeability of a membrane, $P$, which is defined by the
following empirical equation,

$$J = -P \Delta c$$  \hspace{1cm} (4)$$

where $J$ is the flux density of the solute molecules through the membrane and $\Delta c$ is their concentration difference across the membrane.

A membrane is called *selectively permeable* when $P$ depends on the solute being filtered. Our model membrane is selectively permeable to polymer solutes of different lengths as will be shown below and this is the basis for our proposal that tethered membranes be used as a dialysis tool.

From Fick’s law we can derive $P$ in terms of quantities which are more easily measurable, the diffusion constants.

$$\frac{1}{P} = \int_{\text{entanglement zone}} \frac{dz}{D_z(z)}$$  \hspace{1cm} (5)$$

where $D_z$ is a diffusion constant in the perpendicular direction to the membrane and the thickness of the entanglement zone is of order $R_g$, as described earlier. The diffusion constant in this entanglement zone should vary due to the spherical shape of the polymer, as indicated by the $z$-dependent $D_z$ in Eq. (5). But we neglect this as in the previous section and define the average diffusion constant along the $z$-direction, $\bar{D}_z$, using the following formula

$$\frac{R_g}{\bar{D}_z} \equiv \int \frac{dz}{D_z(z)}$$  \hspace{1cm} (6)$$

Then from the definition of $\tau_m$ we get

$$\bar{D}_z = R_g^2/\tau_m \sim N^{-\frac{3}{2}}$$  \hspace{1cm} (7)$$

Hence from Eq. (5) or

$$\frac{1}{P} \approx \frac{R_g}{\bar{D}_z}$$  \hspace{1cm} (8)$$

we get

$$P \sim N^{-2}$$  \hspace{1cm} (9)$$

5 Diffusion constants of a polymer

Now we consider the multiple membrane case. We stack membranes along the $z$-direction with a spacing $d$. A linear polymer of polymerization index $N$ is introduced at an arbitrary
position in this system and the diffusion constants are measured both along the \( z \)-direction and in the \( xy \)-plane using the following equations.

\[
D_z = \lim_{t \to \infty} \frac{\langle (Z(t) - Z(0))^2 \rangle}{2t}
\]

and

\[
D_{xy} = \lim_{t \to \infty} \frac{\langle (X(t) - X(0))^2 + (Y(t) - Y(0))^2 \rangle}{4t}
\]

where \( X, Y \) and \( Z \) are center of mass coordinates of the chain.

First we expect that there should be a crossover in the \( D \)'s at \( d_c \sim R_g \). When \( d \) is smaller than \( d_c \) the chain always entangles with multiple membranes and we expect that some sort of modified reptation should occur. On the other hand when \( d \) is much larger than \( d_c \) the membranes act as small perturbations to the chain dynamics which should be otherwise Rouse-like. We study both limits in turn.

5.1 \( d \ll d_c \)

In this dense regime of membranes we expect the reptation theory to be useful in describing the dynamics of a linear polymer, since the array of closely placed membranes should not be much different from a three-dimensional network.

We first estimate the density of entanglement points. The polymer intersects of order \( R_g/d \sim N^{1/2}b/d \) membranes on average. Furthermore it crosses each membrane of order \( R_g/a \sim N^{1/2}b/a \) times, which can be seen by rescaling the monomer size \( b \) to the mesh size \( a \), so that \( N \to N b^2/a^2 \). Hence the total number of entanglement points is \( R_g^2/ad \sim N b^2/ad \). Then the average number of monomers between entanglement points is obtained by dividing \( N \) by this number, that is, we obtain \( N_e \sim ad/b^2 \). Naive application of reptation theory\[16\] as in section 3 yields

\[
\tau = \tau_1 \frac{N^3b^2}{ad},
\]

\[
D = D_1 \frac{ad}{N^2b^2}
\]

The diffusion constant \( D \) increases as a linear function of the spacing \( d \).
The simulation results for $D_z$ and $D_{xy}$ for $N = 50$ are plotted in Fig. 2 against $d$ up to $d = 30$. Each point in this figure was averaged over 1000 Monte Carlo runs, each spanning longer than the penetration time. As the above argument based on the reptation theory predicts, diffusion should be isotropic in this regime. As Fig. 2 shows, this is indeed the case in the limit of $d/R_g \to 0$. But the system becomes anisotropic rather rapidly as the spacing increases. While $D_z$ remains linear in $d$ even well above $d_c$, $D_{xy}$ deviates dramatically from a straight line around a relatively small $d$.

The scaling behavior in this regime is shown in Fig. 3 and Fig. 4 for chain lengths up to 80. As $d$ increase beyond $d_c$ the scaling clearly breaks down, which is the topic of the next subsection.

5.2 $d \sim d_c$

In this regime the chain diffuses in a layered structure of free space and membrane. Thus depending on the relative importance of each, the dynamics of the chain can be either more Rouse-like or more reptation-like.

We can use a simple resistor-network analogy[18] to derive the effective diffusion constants in this regime. As will be shown shortly $D_z$ and $D_{xy}$ have very different behavior.

First we start with $D_z$. We divide the space into two categories; an entanglement zone of thickness $R_g$ and a Rouse zone of thickness $d - R_g$. The effective resistance per length $d$ in the $z$-direction per unit ($xy$-) area, $\mathcal{R}$, is the sum of two components,

$$\mathcal{R} = \mathcal{R}_E + \mathcal{R}_R$$

or

$$\frac{d}{D_z} = \frac{R_g}{D_E} + \frac{d - R_g}{D_R}$$

where $D_R$ is the Rouse diffusion constant and $D_E \sim N^{-3/2}$ is the average diffusion constant in the entanglement zone as obtained in earlier sections. Then we obtain

$$D_z = D_R \left( 1 + \frac{R_g}{d} \frac{(D_R - D_E)}{D_E} \right)^{-1}$$
Note that the crossover point from the entangled to the Rouse behavior is

\[ d \approx \frac{R_g D_R}{D_E} \sim Nb \] (17)

contrary to the naive expectation of \( N^{1/2}b \).

The diffusion constant in the \( xy \)-plane, \( D_{xy} \), can be obtained in a similar manner except that the two components divided as above are connected in parallel rather than in series.

The effective conductance per unit length along a direction perpendicular to the \( z \)-axis, per rectangular area with height \( d \) and unit width, denoted by \( C \), is again the sum of two components,

\[ C = C_E + C_R \] (18)

or

\[ dD_{xy} = R_g D_E + (d - R_g)D_R \] (19)

yielding

\[ D_{xy} = D_R \left( 1 - \frac{R_g}{d} \frac{D_R - D_E}{D_R} \right) \] (20)

The entangled dynamics cross over to Rouse dynamics at \( d \sim R_g \) differing from the case of \( D_z \) above. Hence in the spacing range \( N^{1/2}b < d < Nb \), we have very anisotropic diffusion constants. While the dynamics in the \( xy \)-plane is almost free the effects of the membranes are still strong along the \( z \)-direction. Beyond this spacing \( \sim Nb \), the dynamics becomes isotropic again with Rouse-like diffusion constants in both directions. The trend is clear from Fig. 2.

The permeability for a stack of \( m \) membranes can be defined in a similar way as in section 4. The permeability is the diffusion constant perpendicular to the slab divided by its effective thickness as shown in Eq. (8). Since the thickness of the stack of \( m \) membranes is \( md \) for large \( m \), the permeability \( P \) is given by

\[ P \sim \frac{D_z}{md} \] (21)

For large \( m \), \( P \) varies as \( N^{-2} \) in the dense regime and \( N^{-1} \) in the dilute regime identical to the scaling of \( D_z \).
6 Escape of an entangled chain from a membrane

So far we have concentrated on the diffusion of the center of mass of a chain. Now we look at the mean square displacements of a monomer in the middle of the chain, which reveals small-scale dynamics in more detail\[19\].

First we place a chain and a membrane so that the chain is maximally entangled, that is, the center of mass of the chain lies in the membrane. After the escape time (defined in section 3), the chain will diffuse from the entanglement zone into free space. This process is shown in Fig. 5 for \(N = 200\). First we see \(t^{1/2}\) behavior up to \(N\tau_1\) which should be the local relaxation of Rouse segments, that is, the chain segment relaxation between entanglement points. At greater times the membrane starts to restrict the motions of chain which makes the relaxation much slower, probably proportional to \(t^{1/4}\) as shown in the figure. In this regime the dynamics in the \(z\)-direction and \(xy\)-direction are different as expected because of the two-dimensional nature of the membrane. We need to simulate larger chains to obtain the fine structure more accurately in this regime.

After the Rouse time scale \(N^2\tau_1\), the chain starts to escape from the membrane. This process is unusually fast in the sense that the exponent is larger than 1, about 3/2 as shown in Fig. 5. This is probably because the farther the chain moves from the membrane the less entangled it is with the membrane, and hence the larger the diffusion constant. The chain will eventually escape from the membrane at a time scale \(N^{5/2}\tau_1\) as obtained in section 3 and normal diffusion \(\langle Z^2 \rangle \sim t\) will resume, which is not shown in the figure.

The monomer relaxation behavior for densely packed multiple membrane case can be interpolated from the data shown here and those given by reptation theory in a three-dimensional network. See, for example, reference \[19\].

7 Conclusion

The dynamics of a flexible polymer in the vicinity of two-dimensional networks of topological obstacles shows interesting behavior. We have investigated the diffusion constants of a polymer of polymerization index \(N\) in a solution in which the two-dimensional networks are
stacked with uniform spacing $d$.

Three regimes of dynamics have been shown to exist as a function of $N$ and $d$.

**Reptation regime** When $d$ is much smaller than the radius of gyration of the polymer $N^{1/2}b$, the dominant mode of relaxation is reptation and simple application of de Gennes’s reptation theory reproduces our Monte Carlo data quite well. The diffusion constant is proportional to $N^{-2}$ and to $d$.

**Intermediate regime** While the chain is almost free in the direction parallel to the membrane when $d > N^{1/2}b$, the motion of the chain perpendicular to the membrane is still dominated by membrane penetration up to $d \sim Nb$. Hence the dynamics of the chain is highly anisotropic in this regime, the ratio $D_{xy}/D_{z}$ being $R_g D_R/(d D_E) \sim N/d$.

**Rouse regime** When $d$ becomes larger than the chain length $Nb$ the diffusion of the chain is predominantly Rouse-like and the correction to the Rouse diffusion constant is $O(N^{\infty}/\epsilon R_g/\lVert \rVert)$ for $D_z$ and $O(R_g/\lVert \rVert)$ for $D_{xy}$. The penetration time for a single isolated membrane is found to be proportional to $N^{5/2}$. This can be interpreted as slowing down of the diffusion of a polymer in the perpendicular direction to the membranes by an extra time of $N^{1/2}\tau_1$ per membrane.

This study in principle can provide a new way of filtrating polymers using highly permeable membranes. The permeability for a single membrane has been found to be proportional to $N^{-2}$. The permeability for multiple membranes will be roughly proportional to $D_z$ divided by the thickness of the stack, providing very rich behavior depending on the spacings between membranes.

Even though we hope that this study gives impetus to filtration techniques using fishnet-like membranes, this work omits important effects. For example we have completely neglected excluded volume effects, which could, for instance, make the membranes effectively repulsive to the polymers. The effect of membrane fluctuations[20, 21, 22] were also neglected in this work. Even though the time scale of large length scale fluctuations of a two-dimensional system is much larger than that of linear polymer relaxation, it could change some important
characteristics of the filtration process, for instance by varying the distances between the membranes, which were assumed uniform in this paper.

Acknowledgment

This work is supported by NSF Grant DMR-9419362 and acknowledgment is made to the Donors of the Petroleum Research Fund, administered by the American Chemical Society for partial support of this research.

References

[1] Yacov Kantor, Mehran Kardar, and David R. Nelson. Statistical mechanics of tethered surfaces. *Phys. Rev. Lett.*, 57(7):791–794, 1986.

[2] E. Guitter, F. David, S. Leibler, and L. Peliti. Thermodynamical behavior of polymerized membranes. *J. Phys. France*, 50:1787–1819, 1989.

[3] Irena B. Petsche and Gary S. Grest. Molecular dynamics simulation of the structure of closed tethered membranes. *J. Phys. I France*, 1:1741–1754, 1993.

[4] Reinhard Lipowsky. Statistical physics of flexible membranes. *Physica A*, 194:114–127, 1993.

[5] Helmut Ringsdorf, Bernhard Schlarb, and Joachim Venzmer. Molecular architecture and function of polymeric oriented systems: Models for the study of organization, surface recognition and dynamics of biomembranes. *Angew. Chem.*, 27:113–158, 1988.

[6] M. Olvera de la Cruz, J.M. Deutsch, and S.F. Edwards. Electrophoresis of a polymer in a strong field. *Phys. Rev. A*, 33:2047–2055, 1986.

[7] J.M. Deutsch and T.L. Madden. Theoretical studies of DNA during gel electrophoresis. *J. Chem. Phys.*, 90(4):2476–2485, 1988.
[8] S.P. Obukhov and M. Rubinstein. Scaling of megabase DNA undergoing gel electrophoresis. *J. Phys. II France*, 3(10):1455–1459, 1993.

[9] Christoph F. Schmidt, Karel Svoboda, Ning Lei, Irena B. Petsche, Lonny E. Berman, Cyrus R. Safinya, and Gary S. Grest. Existence of a flat phase in red cell membrane skeletons. *Science*, 259:952–955, 1993.

[10] Robert F. Service. Making molecular filters more reactive. *Science*, 265:1363, 1993.

[11] Iwao Teraoka, Kenneth H. Langley, and Frank E. Karasz. Reptation dynamics of semi-rigid polymers in porous media. *Macromolecules*, 25:6106–6112, 1992.

[12] Bruno H. Zimm and Oscar Lumpkin. Reptation of a polymer chain in an irregular matrix: Diffusion and electrophoresis. *Macromolecules*, 26:226–234, 1993.

[13] J.M. Deutsch and Hyoungsoo Yoon. Simulation of forces on entangled polymers due to slippage. To be published in *J. Chem. Phys.*, 1995.

[14] M. Doi. *Polym. J.*, 5:288, 1973.

[15] K.E. Evans and S.F. Edwards. Computer simulation of the dynamics of highly entangled polymers: Part 1 equilibrium dynamics. *J. Chem. Soc. Faraday Trans. II*, 77:1891–1912, 1981.

[16] Pierre-Gilles de Gennes. *Scaling Concepts in Polymer Physics*. Cornell University Press, 1979.

[17] Sergei P. Obukhov, Michael Rubinstein, and Thomas Duke. Dynamics of a ring polymer in a gel. *Physical Review Letters*, 73(9):1263–1266, 1994.

[18] M. Daoud and P.G. de Gennes. Some remarks on the dynamics of polymer melts. *J. Polym. Sci. Phys ed.*, 17:1971–1981, 1979.

[19] M. Doi and S.F. Edwards. *The Theory of Polymer Dynamics*. Clarendon Press, 1986.

[20] Joseph A. Aronovitz and T.C. Lubensky. Fluctuations of solid membranes. *Phys. Rev. Lett.*, 60(25):2634–2637, 1988.
[21] Reinhard Lipowsky and Marc Girardet. Shape fluctuations of polymerized or solidlike membranes. *Phys. Rev. Lett.*, 65(23):2893–2896, 1990.

[22] Farid F. Abraham and David R. Nelson. Fluctuations of the flat and collapsed phases of polymerized membranes. *J. Phys. France*, 51:2653–2672, 1990.
Figure 1: The logarithm of penetration time, $\tau$, defined in the text is plotted against the logarithm of the polymerization index, $N$. The membranes are placed at distances 5, 6, 8, 10, 12, 14 and 18 for $N = 8, 10, 16, 24, 36, 50$ and 80. The distances are chosen so that the probability of the chain touching two membranes at the same time is extremely small.
Figure 2: The diffusion constants are given as functions of $d$ for $N = 50$. The circles represent $D_{xy}$ and the triangles $D_z$. The Rouse value is around $5.5 \times 10^{-3}$ in our unit. Each pair of points represents an average over 1000 Monte Carlo runs.
Figure 3: Scaling graph for $D_z$ for $N = 10$(open triangle), 16(cross), 24(pentagon), 36(filled triangle), 50(snowflake) and 80(circle). The scaling function is $D_z = D_1/N^{1.5} F(d/R_g)$ where $R_g = \sqrt{Nb^2/6}$ is used. $F$ is pretty linear when $d/R_g$ is small and the scaling breaks down beyond that. Note that each data point was obtained separately. Each point is an average of 1000 Monte Carlo runs.
Figure 4: Scaling graph for $D_{xy}$. The symbols represent the same $N$’s as in Fig. 3. The break-down of the scaling when $d > R_g$ is more dramatic here than in Fig. 3.
Figure 5: Average mean square displacements of 40 monomers at the center of a chain with $N = 200$. $r_i$ represents $z$-component (triangle) or $x$ (or $y$)-component (circle) of the coordinate of a monomer. The curves were averaged over 200 Monte Carlo runs. After the $t^{1/4}$ regime $D_z$ seems to show $t^{1/2}$ behavior again, but it is not very clear from this graph.