



Theoretical Study of Adhesion Fluids Membranes with Polymer Chains Grafted

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Abstract—The system that we consider is two fluid membranes that are decorated with long flexible polymers. We are interested in a physical system consisting two adjacent membranes with end-grafted (or adsorbed) polymers. In addition to pure interactions between membranes, the presence of polymers gives rise to new induced mediated repulsive interactions. In fact repulsive interactions are caused by the excluded-volume forces between grafted polymers. In this work, we propose ourselves to study the effects caused by the polymer chains grafted on the mutual adhesion of two adjacent fluid membranes. First of all, we will establish any kind of interactions which comes into play. Then, we give in detail the thermodynamics of the transition of separation in the two regimes. Finally, we will determine the median value of the distance of separation of the system.

Keywords—Adhesion, Biological membranes, Polymer-tethered.

I. INTRODUCTION

The cellular adhesion is a phenomenon indispensable that is implied in most physiological cellular phenomena (survival, differentiation, migration, activation) and of the pathological situations (formation of cancerous metastases, tissue invasion by a pathogen, inflammation, reaction of the organization with respect to the biomaterials) (e.g. [1]–[11]).

During last decades, the progress of immunology (immunology and the study of the immune system are essential tools for two particular domains: the rejection of Clerc's Office and autoimmune diseases, like the diabetes), and of the molecular genetics allowed to characterize hundreds of receptors of adhesion. In fact, it is possible that about half of the molecules present on the biological membrane of a cell are implied in adhesion.

However, a lot of experiments showed that it was not enough to identify the molecules of adhesion present on two biological surfaces to predict if their meeting would lead to an attachment. In another domain, the cadherins (or CDH) are a class of glycoproteins that are expressed on the cellular surface.

They play important roles in the cellular adhesion with the result that they ensure the intercellular connection within tissues), are homotypic molecules of adhesion pretence able to produce a segregation of cellular populations. However, the mechanisms of this selectivity are difficult to explain qualitatively insofar as interactions were described between different molecules of cadherins.

Moreover, to start a disease, the infectious bacteria must initially penetrate in the organism and adhere to tissue. The Factors of adhesion allow the fixing of the bacteria in a cell. The invasive power is the capacity of the bacterium to be spread and to multiply in tissues. Let us note that there exist several prerequisites, in particular to carry out and control adhesive surfaces definite on a subnanometric scale, and to understand the Brownian movement of a particle in the vicinity of a wall. For this purpose, we propose ourselves to associate with the traditional tools of the cellular biology of the concepts, the physical methods necessary for measurement and the exploitation of such a phenomenon of adhesion.

In this work, we propose ourselves to study the effects caused by the polymer chains grafted on the mutual adhesion of two adjacent fluid membranes. First of all, we will establish any kind of interactions which comes into play. Then, we give in detail the thermodynamics of the transition of separation in the two regimes. finally, we will determine the median value of the distance of separation of the system which depends on an external constraint which is the multiplier of Lagrange.

II. GENERIC INTERACTIONS

Consider two fluid membranes in the presence of long flexible polymer chains. For simplicity, these chains are assumed to have the same polymerization degree, N . we indicate ourselves by Γ . The grafting density, is the number of grafted polymer chains per unit area. In addition of these two variables Γ and N , the properties of a chain depend on the parameter of excluded volume v . Then, we are concerned with a monodisperse system. We denote by $\Gamma^* \sim R_F^{-2} \sim a^{-2} N^{-6/5}$ the critical grafting density who separates two regimes: the mushrooms regime ($\Gamma < \Gamma^*$) and the brush regime ($\Gamma > \Gamma^*$). Here R_F represent the Flory radius and a denotes the monomer size. Thus, the thermodynamic transition depends strongly on the value of the density of grafting.

In this work, The main goal is a quantitative study of the unbinding transition thermodynamics that is drastically affected by the associated polymers.

For fluid membranes with end-grafted (or adsorbed) polymers, Their cohesion is provided by long-ranged van der Waals forces(e.g.[12]). But these attractive interactions are balanced, at short membrane separation, by strong repulsion coming from hydration forces for uncharged bilayers (e.g.[13]).In addition, for bilayers carrying electric charges, the attractive interaction is reduced by the presence of electrostatic forces(e.g.[12, 14, 15]).

Suppose, now, that the two adjacent membranes are decorated by many long polymer chains. For strong grafting density, the two bilayers can be considered such as soft brushes. When the two bilayers approach one of the other, at a certain distance, the polymer brushes begin to overlap, and thus, the monomer concentration increases. Therefore, the osmotic pressure increases and leads to a repulsive force(e.g.[16]–[17]).

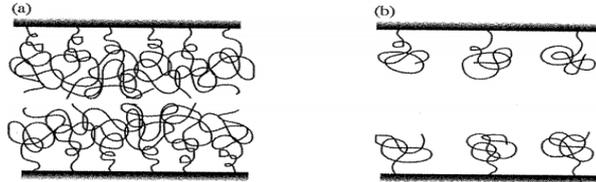


Figure.1 Polymer chains grafted at a planar interface : (a) brushes, at high grafting density, (b) mushrooms, at low grafting density .

It comes by the excluded-volume interactions between monomers that we discuss later. In this context, Milner, Witten and Cates(e.g.[18]). used a more realistic profile for the density of polymers and obtained the following form of the interaction energy per unit area(e.g.[19]).

$$V_P(l) = k_B T L_0 \Gamma \left(\frac{\pi^4 v \Gamma}{144 a^4} \right)^{1/3} \left[\left(\frac{l}{2L_0} \right)^2 + \frac{2L_0}{l} - \frac{1}{5} \left(\frac{l}{2L_0} \right)^5 - \frac{9}{5} \right] . \quad (1)$$

(The index P is used to designate the polymer). Here l is the distance between two bilayers, a_m is the size of the monomer, $v_m = a_m^3 (1 - 2\chi)$ represent the excluded-volume parameter, χ is the Flory–Huggins interaction parameter. Withan athermal solvent, for which intramolecular interactions other than steric may be neglected $v_m \approx a_m^3$. Hristova and Needham(e.g.[20])find that for the oxyethylenemonomer unit of (PEG) Poly(ethyleneglycol): $a_m \approx 0.35 \text{ nm}$. From adhesionmeasurements, Evans et al. (e.g.[21])find that the data for (PEG) lipids is best fitted by a value of $a_m \approx 0.43 \text{ nm}$. These values close to the size of an oxyethylene unit $a_m \approx 0.39 \text{ nm}$ determined from the monomer volume in aqueous solution(e.g.[21]),and

$$L_0 = 1,07 N (v a^2 \Gamma)^{1/3} \quad (2)$$

denotes the size of the grafted layer. Of course, the above expression of the potential remains valid for $l < 2L_0$. For $l > 2L_0$ this potential vanishes.

Let us recall that another expression of the interaction energy was introduced by de Gennes[22, 23]. The author used a scaling approach to establish the density profile of a brush polymer. It found the following expression

$$V_P(l) = \frac{8}{35} k_B T L_0 \Gamma^{3/2} \left[7 \left(\frac{2L_0}{l} \right)^{5/4} + 5 \left(\frac{l}{2L_0} \right)^{7/4} - 12 \right] , \quad (3)$$

with the new size of the grafted layer

$$L_0 = N a^{5/3} \Gamma^{1/3} . \quad (4)$$

The repulsive pressure developed between the surfaces can be calculated from the de Gennes equation

$$\Pi_P(l) = k_B T \Gamma^{3/2} \left[\left(\frac{2L_0}{l} \right)^{9/4} - \left(\frac{l}{2L_0} \right)^{3/4} \right] , \quad l < 2L_0 . \quad (5)$$

The first term in Eq. (5) arises from the osmotic repulsion between the coils which favors their stretching, and the second term is due to the elastic energy of the chains which opposes stretching.

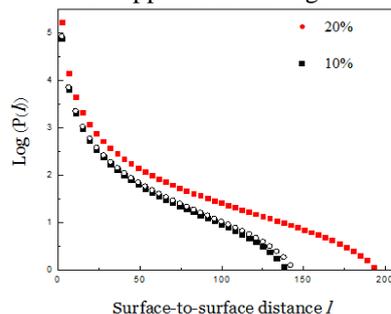


Figure 2 : Osmotic pressure P versus fluid spacing between opposing lipopolymers-grafted bilayers. The solid curve are fits to the grafting mol fraction $f = 0.1$ and $f = 0.2$ data using Eq. (5)

Example: The surface of a material is covered with a water-soluble nonionic polymeric surfactant such that the surface concentration of the surfactant is $1 \times 10^{-6} \text{ mol} / \text{m}^2$. We Calculate the repulsive pressure at 298 K when two such surfaces are brought close to each other such that $l/2L_0 = 0.9$.

Therefore, from Eq. (5) we get,

$$\Pi_p(l) = \frac{1,38 \times 10^{-23} \times 298}{(1,29 \times 10^{-9})^3} \left[\left(\frac{1}{0,9} \right)^{9/4} - (0,9)^{3/4} \right] = 6,6 \times 10^5 \text{ Pa} \quad (6)$$

Let us notice that the latter is the same thickness of brush as that reported by Milner, Witten, and Cates, by supposing that $v = a^3$. The expression (3) is largely used for made experimental data, because it depends only on two parameters ρ and L_0 , who have a physical sense.

For very small distances compared to the brush thickness, the previous expression of the potential of interaction, is reduced to

$$V_p(l) = \frac{56}{35} k_B T L_0 \Gamma^{3/2} \left(\frac{2L_0}{l} \right)^{5/4}, \quad l \ll 2L_0 \quad (7)$$

It should be noted that all the expressions described above are applicable to high density of grafting. For the low density of grafting, characterized by $\Gamma < \Gamma^* \sim a^{-2} N^{-6/5}$ (mushroom regime), Dolan and Edwards (e.g.[24]). Are calculated the energy of repulsive interaction per unit area in a good solvent and between two surfaces covered with polymers

$$V_p = k_B T \Gamma \left[\ln \left(\frac{l}{4\sqrt{\pi} R_F} \right) + \frac{\pi^2 R_F^2}{l^2} \right], \quad l \leq 3\sqrt{2} R_F \quad (8a)$$

$$V_p = k_B T \Gamma \ln \left(1 - 2e^{-l^2/4R_F^2} \right), \quad l > 3\sqrt{2} R_F \quad (8b)$$

in particular, at very small distances from the radius of gyration, we have

$$V_p = k_B T \Gamma \pi^2 \frac{R_F^2}{l^2}, \quad l \ll R_F \quad (9)$$

The steric repulsion between the solid substrates was measured directly by using both the surface forces[25, 26] and atomic force microscope[27, 28]. The steric forces between the vesicles, is also observed by Kenworthy and al.(e.g.[29]). using the method of osmotic pressure. Experimentally, Milner, Witten, Cates and the model of de Gennes, reproduce quite well the force curves between polymer brushes in a good solvent.

For two parallel bilayer membranes that are a finite distance l apart, the total interaction energy (per unit area) is the following sum

$$V_T(l) = V_{HW}(l) + V_{SP}(l) \quad (10)$$

Where

$$V_{HW}(l) = V_H(l) + V_W(l) \quad (11)$$

The first part represents the hydration energy. The hydration forces that act at small separation of the order of 1 nm, have been discovered for multilayers under external stress(e.g.[30]-[34]). The adopted form for the hydration energy is an empirical exponential decay

$$V_H(l) = A_H e^{-l/\lambda_H} \quad (12)$$

The typical values of amplitude A_H and potential-range λ_H are $A_H \sim 0.2 \text{ J/m}^2$ and $\lambda_H \sim 0.3 \text{ nm}$.

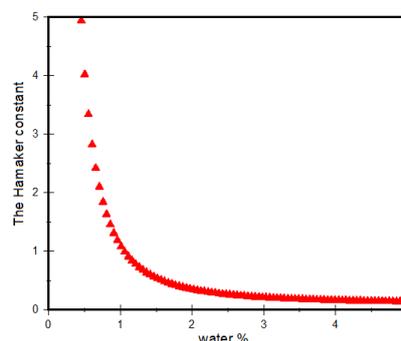


Figure 3. The Hamaker constant between polymer/water layers interacting across water as a function of water content of the adsorbed layer.

Tableau 1-Parametervalue of effective interaction between membranes “T.Charitat,S.Lecuyer et al³⁵, And H.I.Petrache, N.Gouliaev et al³⁶ (DSPC multilayer).

Ref	System	T(°C)	A(×10 ⁻²¹ J)	P(×10 ⁶ Pa)	d (nm)
[35]	DSPC	43	0	27	1.3
[35]	DSPC	53	1.	29.5	1.3
[36]	DPPC	50	2.87-9.19	13.8-19.5	1.97-2.39

The second part is the attractive van der Waals energy that results from polarizabilities of lipid molecules and water molecules. This interaction energy has the standard form

$$V_w(l) = -\frac{W}{12\pi} \left[\frac{1}{l^2} - \frac{2}{(l+\delta)^2} + \frac{1}{(l+2\delta)^2} \right] \quad (13)$$

The Hamaker constant is in the range $W \sim 10^{-21} - 10^{-22}$ J. In the above expression, the bilayer thickness δ is of the order of $\delta \sim 4$ nm.

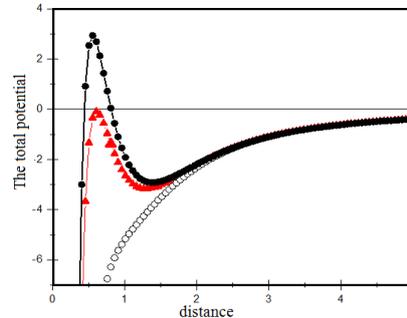


Figure 4: The total potential as function of distance for three cases [32]

The undulation force, which is important for controlling the swelling behavior of the lamellar stack, is a long-range force generated by the out-of-plane bilayer fluctuations and it is therefore of steric and entropic origin. Helfrich [35] showed by simple scaling arguments that the free-energy density, $V_s(l)$, which leads to undulation forces for fluid membranes, takes the form

$$V_s(l) = C_H \frac{(k_B T)^2}{\kappa l^2} \quad (14)$$

With k_B the Boltzmann's constant, T the absolute temperature, l is the mean distance between layers, and κ the common bending rigidity constant of the two membranes. But, for two bilayers of different bending rigidity constants κ_1 and κ_2 , we have $\kappa = \kappa_1 \kappa_2 / (\kappa_1 + \kappa_2)$. The exact value of the prefactor C_H is unknown, but estimates have been derived from crude theoretical approximations by Helfrich $C_H \sim 0.23$ [28], but computer simulations predict smaller prefactors, namely, $C_H \sim 0.16$ [36], $C_H \sim 0.1$ [37], $C_H \sim 0.07$ [38] and $C_H \sim 0.08$ [39]. Therefore, the steric interaction energy is significant only for those membranes of small bending modulus. Of course, this energy vanishes for rigid interfaces ($\kappa \rightarrow \infty$).

The following paragraph is devoted to study the Unbinding transition of two bilayer membranes with grafted polymers chains.

III. THE UNBINDING TRANSITION

We note that the lamellar phase remains stable at the minimum of the potential, provided that the potential-depth is comparable to the thermal energy $k_B T$. This depends, in particular, on the value of W -amplitude.

From a theoretical point of view, Lipowsky and Leibler [40] had predicted a phase transition that drives the system from a state where the membranes are bound to a state where they completely separate. Such a phase transition is first-order if the steric repulsive energy is taken into account. But, if this energy is ignored (for relatively rigid membranes), the transition is rather second-order. We restrict ourselves to second-order phase transitions, only. The authors have shown that there exists a certain threshold W_c beyond which the van der attractive interactions are sufficient to bind the membranes together, while below this characteristic amplitude, the membrane undulations dominate the attractive forces, and then, the membranes separate completely. In fact, the critical value W_c depends on the parameter of the problem, which are temperature T , and parameters A_H , λ_H , δ and δ . For room temperatures and $A_H \sim 0.2$ J/m², $\lambda_H \sim 0.3$ nm and $\delta \sim 4$ nm, one has $W_c \sim (6.1 - 0.61) \times 10^{-21}$ J, when the bending rigidity constant is in the range $\kappa \sim (1 - 20) \times 10^{-19}$ J. For instance, for egg lecithin, one has [63] $\kappa \sim (1 - 2) \times 10^{-19}$ J, and the corresponding threshold W_c is in the interval $W_c \sim (6.3 - 4.1) \times 10^{-21}$ J, We note that the typical value W_c corresponds to some temperature, T_c , called unbinding critical temperature [40; 42].

Let us first consider uncharged membranes, and notice that the Hamaker constant may be varied changing the polarizability of the aqueous medium. It was found [40] that, when the critical amplitude is approached from above, the mean-separation between the two membranes diverges according to

$$\langle l \rangle \sim (T_c - T)^{-\psi} \sim (W - W_c)^{-\psi}, \quad (W \rightarrow W_c^+ \text{ or } T \rightarrow T_c^-) \quad (15)$$

Here, ψ is a critical exponent whose value is [40]

$$\psi \sim 1.00 \pm 0.03 \quad (16)$$

Such an exponent was computed using field-theoretical Renormalization-Group.

From an experimental point of view, critical fluctuations in membranes were considered in some experiment [43], and in particular, the mean-separation was measured.

We said above that, for $T > T_c$, the shape fluctuations drive the membranes forming lamellar phase apart even in the presence of the direct attractive forces. In this case, the system recovers its bound state by a simple application of an external pressure or a lateral tension.

In the presence of an external pressure P , it was found [42] that the mean-separation scales as :

$$\langle l_0 \rangle = 2^{1/3} (K_B T)^{2/3} \kappa^{-1/3} P^{-1/3} \quad (17)$$

We ignored a numerical factor of the order of unity. This behavior is in agreement with the MC data. [42] Naturally, the study of the phenomenon of adhesion depends critically on the value of the grafting density

IV. MUSHROOM REGIME

Note, first, that for small separations compared to the radius of gyration, the energy of steric interaction is defined by Eq. (9). Note that this shape is similar to that of Helfrich, relation (14). Thus, for these distance scales, the sum $V_s + V_p$ reduces to

$$V_{SP}(l) = V_s(l) + V_p(l) = c_H \frac{(k_B T)^2}{\hat{\kappa} l^2}, \quad l < R_F, \quad (18)$$

where the parameter $\hat{\kappa}$ is given by

$$\hat{\kappa}^{-1} = \kappa^{-1} + \frac{\pi^2}{c_H k_B T} \left(\frac{\Gamma}{\Gamma^*} \right). \quad (19)$$

We used the following formula $\Gamma R_F^2 = \Gamma / \Gamma^*$. The expression (19) informs us that, in the low grafting density regime, the chains have grafting effect to modify the energy fluctuations of form through a renormalized additional bending modulus. Therefore, the effective bending modulus $\hat{\kappa}$ is smaller than κ departure. This means that the critical parameter separation increased by the presence of grafting polymers. We easily show that the critical value W_c^* decorated membranes can be connected to the common by

$$\frac{W_c^*}{W_c} = \left(\frac{\kappa}{\hat{\kappa}} \right)^{1/2} = \left[1 + \frac{\kappa \pi^2}{c_H k_B T} \left(\frac{\Gamma}{\Gamma^*} \right) \right]^{1/2}. \quad (20)$$

As it should, the critical value of the amplitude- W increases by the presence of the forces of excluded volume. The report above, depends only on the density grafting scaled down Γ / Γ^* and the dimensionless bending modulus $\kappa / k_B T$.

In summary, in the mushroom regime, the average separation $\langle l \rangle$ can be given by (15) or (17) by replacing κ and W_c by the parameters effective corresponding $\hat{\kappa}$ and W_c^* . In particular, when the pressure P is applied we have

$$\frac{\langle l \rangle}{\langle l \rangle_0} = \left(\frac{\kappa}{\hat{\kappa}} \right)^{1/3} = \left[1 + \frac{\kappa \pi^2}{c_H k_B T} \left(\frac{\Gamma}{\Gamma^*} \right) \right]^{1/3}. \quad (21)$$

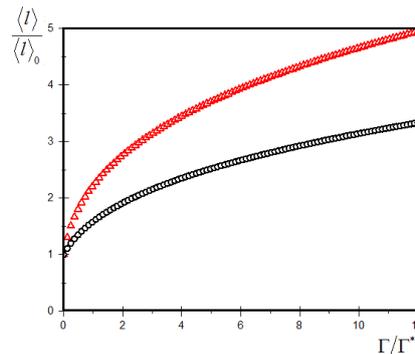


Figure 5: the average distance as a function grafting density Γ / Γ^* .

V. BRUSH REGIME

In this regime, a small distances compared to the total layer grafting $2L_0$. The steric interaction energy is defined by Eq. (7). We start the discussion by asking

$$\Delta F_1(l) = \frac{56}{35} k_B T L_0 \Gamma^{3/2} \left(\frac{2L_0}{l} \right)^{5/4} - \frac{W}{12\pi l^2} \quad (22)$$

This means that we want to compare the steric energy by the van der Waals forces. It is easy to see that $\Delta F_1(l)$ is not limited from below ($\Delta F_1(l) = -\infty$) and vanishes at some point, and has a maximum. Thus, the interaction energy is not stable. In addition, the interaction tends to remove the corrugations of the membrane. Therefore, in the brush regime the only way to connect the two bilayers is repulsive to exert pressure perpendicular P_{\perp} . To analyze the bending phenomenon, we consider the sum

$$\Delta F_2(l) = Pl + \frac{56}{35} k_B T L_0 \Gamma^{3/2} \left(\frac{2L_0}{l} \right)^{5/4} \quad (23)$$

Note that the Lagrange multiplier P can be interpreted as current necessary pressure to fix the value of the separation between the bilayers a certain value average $\langle l \rangle$. By minimizing the energy with respect to the separation is obtained

$$\langle l \rangle = 2L_0 \Gamma^{3/2} \left(\frac{P}{k_B T} \right)^{-5/9} \quad (24)$$

This expression must be compared to that on the pure lamellar phase, which decreases more as $P^{-1/3}$.

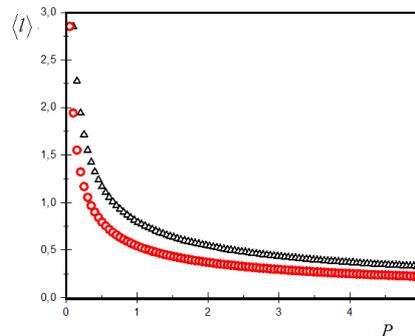


Figure 6: The average distance as a function of pressure

Figure (6) show that the higher the pressure increases the average distance becomes less important, and after the relationship (22), the average distance also depends on the temperature and pressure for a fixed average distance becomes inversely proportional to the temperature

VI. CONCLUSIONS

We recall that the aim of this work was to study the effects caused by polymer chains grafted on mutual adhesion of two fluid membranes. To do this, we first recalled the expressions of the energy of steric interaction, according mushroom and brush regimes. In each case of figure, we have studied in detail the thermodynamics of the transition of Accession which causes the system with a separate lamellar phase to a phase where the two adjacent membranes are related.

In this work, we have evidence generic interactions between the lipid bilayers forming the lamellar phase. In the first place, we have given the expressions of steric energy in two regimes: the mushroom regime and the brush regime. Then we set the total energy interaction between the two bilayers separated by a distance l denies. This energy is a contribution of a number of terms such as energy of hydration interaction, van der Waals interaction energy and energy steric interaction. On the other hand, we have studied in detail the thermodynamics of the transition from adhesion phenomenon that puts the system in a lamellar phase where both membranes are completely separated in a phase where the two bilayers remain in contact. For the case of transition of separation we have also established the average value of the distance separating these two bilayers. The latter is directly dependent on an external pressure P is, in fact, a Lagrange multiplier. Note that P is the pressure required to fix the value of the separation distance of the system.

In conclusion, we emphasize the fact that the stability of such a system depends on a number of parameters, ie, temperature, the amplitude of the van der Waals interaction and the external pressure. Note also that these systems into bilayer has an undeniable interest in many areas of biological research, pharmaceutical (drug transport) and the food industry

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