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ORIGINAL CONTRIBUTION

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Electrostatic repulsion between two parallel plates covered with polymer brush layers

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Abstract An expression for the electrostatic repulsive force is obtained for two parallel similar plates immersed in an electrolyte solution at separation h covered with a uniformly charged polymer brush layer of intact thickness d_o under compression ($h < 2d_o$) after the two brushes come into contact. It is assumed that when the two brushes come into contact, they are squeezed against each other but do not interdigitate. The electrostatic repulsive force is found to increase with decreasing h as $1/h$ for highly charged brushes and as $1/h^2$ for weakly charged brushes. This is in contrast to the interaction force between the brush layers before contact ($h \geq 2d_o$), which is essentially proportional to $\exp[-\kappa(h - 2d_o)]$ (where κ is the Debye–Hückel parameter). It is also shown that the interaction force for highly charged brushes, which becomes independent of the electrolyte concentration, can be comparable in magnitude to the steric repulsive forces between the brushes resulting from osmotic repulsion and the elastic energy of the brushes.

Key words Polymer brush layer – Electrostatic repulsion

Introduction

Interaction forces or energies between colloidal particles are considerably affected by coating the particles with polymers [1–14]. When two colloidal particles covered with graft polymers (i.e., polymer brush layers) approach each other, steric repulsion acts between the polymer brush layers on the respective particles. A theory of de Gennes [3] assumes that when the two brushes come into contact, they are squeezed against each other but do not interdigitate. Taunton et al. [5] found that their experimental data on the repulsion between two polystyrene brush layers are in excellent agreement with the theory of de Gennes (see also Ref. [4]). This theory [3] is applied for the steric interaction between uncharged brushes. If brushes are charged, then electrostatic repulsion between brushes must also be taken into account in addition to steric repulsion. In the present paper we extend the model of de Gennes [3] to the case of charged polymer brushes and obtain an expression for the electrostatic repulsion between brushes after they come into contact. We restrict ourselves to the case where the brushes are squeezed against each other but do not interdigitate. We will thus not consider electrostatic interactions for the case where the polymer layers on interacting particles penetrate each other [6].

Theory

Before we consider the electrostatic interaction between two parallel brushes after they come into contact, we first treat the case where the brush layers are not in contact.

Repulsion between intact brushes

Consider two parallel identical plates coated with a charged polymer brush layer of intact thickness d_o at separation h immersed in a symmetrical electrolyte solution of valence v and bulk concentration n as shown in Fig. 1a. We assume that dissociated groups of valence z are uniformly distributed in the intact brush layer at a number density N_o. We first obtain the potential distribution in the system when the two brushes are not in contact ($h > 2d_o$). We take an x-axis perpendicular to the brushes with its origin 0 at the core surface of the left plate so that the region $d_o < x < h - d_o$ is the electrolyte solution and the regions $0 < x < d_o$ and $h - d_o < x < h$ are the brush layers. We also assume that the brush layers are penetrable to electrolyte ions as well as water molecules. As a result of the symmetry of the system we need consider only the regions $d_o < x < h/2$ and $0 < x < d_o$. The Poisson–Boltzmann equations for the electric potential $\psi(x)$ in the respective regions are

$$\frac{\mathrm{d}^2\psi}{\mathrm{d}x^2} = -\frac{\rho_{e1}}{\varepsilon_r\varepsilon_o}, \quad d_o < x < h/2 \ , \tag{1}$$

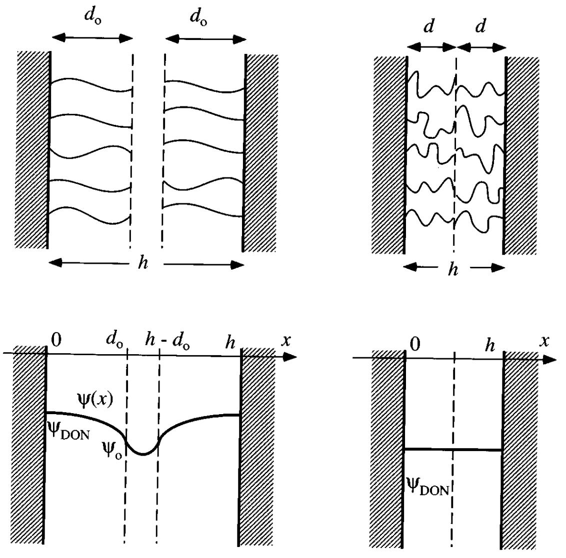
$$\frac{\mathrm{d}^2\psi}{\mathrm{d}x^2} = -\frac{\rho_{e1} + \rho_{\mathrm{fix}}^{(0)}}{\varepsilon_r\varepsilon_o}, \quad 0 < x < d_o \ , \tag{2}$$

where

$$\rho_{\mathrm{fix}}^{(0)} = zeN_o \tag{3}$$

is the fixed charge density in the intact brush layer,

$$\rho_{\mathrm{el}} = ve[n_+(x) - n_-(x)] \tag{4}$$



(a) Before contact

(b) After contact

Fig. 1a, b Interaction between two identical parallel plates covered with a charged polymer brush layer and the potential distribution across the brush layers. **a** Before the two brushes come into contact ($h \geq 2d_o$). **b** After the two brushes come into contact ($h < 2d_o$). d_o is the thickness of the intact brush layer

is the charge density resulting from the electrolyte ions, e is the elementary electric charge, ε_r is the relative permittivity of the solution, and ε_o is the permittivity of a vacuum. If the distribution of the electrolyte ions obeys a Boltzmann law, then

$$n_\pm(x) = ne^{\mp y(x)} \tag{5}$$

and

$$\rho_{\mathrm{el}} = -2ven\sinh[y(x)] \ , \tag{6}$$

with

$$y(x) = \frac{ve\psi(x)}{kT} \ , \tag{7}$$

where k is the Boltzmann constant, T is the absolute temperature, and y is the scaled potential. Substituting Eqs. (3), (6) and (7) in to Eqs. (1) and (2), we obtain

$$\frac{\mathrm{d}^2y}{\mathrm{d}x^2} = \kappa^2\sinh y, \quad d_o < x < h/2 \ , \tag{8}$$

$$\frac{\mathrm{d}^2y}{\mathrm{d}x^2} = \kappa^2(\sinh y - \sinh y_{\mathrm{DON}}^{(0)}), \quad 0 < x < d_o \ , \tag{9}$$

with

$$\kappa = \left(\frac{2v^2e^2n}{\varepsilon_r\varepsilon_o kT}\right)^{1/2} \tag{10}$$

$$y_{\mathrm{DON}}^{(0)} = \frac{ve\psi_{\mathrm{DON}}^{(0)}}{kT} \tag{11}$$

and

$$\sinh y_{\mathrm{DON}}^{(0)} = \frac{zN_o}{2vn} \tag{12}$$

or equivalently

$$y_{\mathrm{DON}}^{(0)} = \ln\left[\frac{zN_o}{2vn} + \sqrt{\left(\frac{zN_o}{2vn}\right)^2 + 1}\right] \ , \tag{13}$$

where κ is the Debye–Hückel parameter and $\psi_{\mathrm{DON}}^{(0)}$ is the Donnan potential of the intact polymer brush layer. Equation (8) is the usual non-dimensional Poisson–Boltzmann equation and the second term on the right-hand side of Eq. (9) comes from the contribution from the fixed charges of density zeN_o in the polymer brush layer. The boundary conditions for $y(x)$ are [7–9]

$$\left.\frac{\mathrm{d}y}{\mathrm{d}x}\right|_{x=h/2} = 0 \tag{14}$$

$$y(d_o^-) = y(d_o^+) \tag{15}$$

$$\left.\frac{\mathrm{d}y}{\mathrm{d}x}\right|_{x=d_o^-} = \left.\frac{\mathrm{d}y}{\mathrm{d}x}\right|_{x=d_o^+} \tag{16}$$

$$\left.\frac{\mathrm{d}y}{\mathrm{d}x}\right|_{x=0^+} = 0 \ . \qquad (17)$$

Equation (14) follows from the symmetry of the system. Equations (15) and (16) state that the potential and its derivative are continuous at the boundary between the brush and in the solution phase. Equation (16) assumes that the relative permittivity has the same value in the brush layer and in the electrolyte solution. Equation (17) implies that the core surface at $x = 0$ is uncharged.

The electrostatic interaction force $P_\mathrm{e}(h)$ per unit area between the two charged brush layers at separation h when they are separated $(H \geq 2d_\mathrm{o})$ is given by the osmotic pressure at the midpoint between the plates $x = h/2$ minus that in the bulk solution phase, viz.,

$$P_\mathrm{e}(h) = [n_+(h/2) + n_-(h/2)]kT - 2nkT \ , \qquad (18)$$

where $n_+(h/2)$ and $n_-(h/2)$ are, respectively, the concentrations of cations and anions at $x = h/2$. Thus we have

$$P_\mathrm{e}(h) = 4nkT \ \sinh^2[y(h/2)/2], \quad h \geq 2d_\mathrm{o} \ . \qquad (19)$$

The solution to Eqs. (8) and (9) subject to the boundary conditions Eqs. (14)–(17) takes a complicated form, involving numerical integration. For the case where $\kappa d_\mathrm{o} \gtrsim 1$ we have previously found that the value of $y(h/2)$ can be calculated by solving the following coupled equations [8]:

$$2\cosh y_\mathrm{DON} + \frac{zN_\mathrm{o}}{vn}[y_\mathrm{o} - y_\mathrm{DON}] = 2\cosh y(h/2) \qquad (20)$$

$$\cosh\left(\frac{y_\mathrm{o}}{2}\right) = \cosh\left(\frac{y(h/2)}{2}\right) \cdot \mathrm{dc}\left[\frac{kh}{2} \cdot \cosh\left(\frac{y(h/2)}{2}\right), 1/\cosh\left(\frac{y(h/2)}{2}\right)\right], \qquad (21)$$

where dc is a Jacobian elliptic function with modulus $1/\cosh[y(h/2)]$ and $y_\mathrm{o} \equiv y(d_\mathrm{o})$ is the scaled unperturbed potential at the front edge $x = d_\mathrm{o}$ of the intact brush layer (i.e., the potential at the boundary between the brush layer and the surrounding electrolyte solution before the two brushes come into contact) and is given by [7–9]

$$\begin{aligned} y_\mathrm{o} &= y_\mathrm{DON}^{(0)} - \tanh\left(y_\mathrm{DON}^{(0)}/2\right) \\ &= \ln\left[\frac{zN_0}{2vn} + \sqrt{\left(\frac{zN_0}{2vn}\right)^2 + 1}\right] + \frac{2vn}{zN_\mathrm{o}}\left[1 - \sqrt{\left(\frac{zN_0}{2vn}\right)^2 + 1}\right] \end{aligned} \qquad (22)$$

For $\kappa d_\mathrm{o} \gtrsim 1$, the potential deep inside the polymer brush layer becomes the Donnan potential $\psi_\mathrm{DON}^{(0)}$. Further, for $\kappa(h - 2d_\mathrm{o}) \gtrsim 1$, we have previously found [8] that $P_\mathrm{e}(h)(h \geq 2d_\mathrm{o})$ is approximated by

$$P_\mathrm{e}(h) = 64nkT \tanh^2(y_\mathrm{o}/4)\exp[-\kappa(h - 2d_\mathrm{o})], \quad h \geq 2d_\mathrm{o} \ . \qquad (23)$$

The potential energy of the electrostatic interaction between the two brushes per unit area is obtained by integrating $P_\mathrm{e}(h)$ with respect to h. If Eq. (23) is adopted, we obtain

$$V(h) = \frac{64nkT}{\kappa}\tanh^2(y_\mathrm{o}/4)\exp[-\kappa(h - 2d_\mathrm{o})], \quad h \geq 2d_\mathrm{o} \ . \qquad (24)$$

Repulsion between compressed brushes

Next we consider the situation after the two brushes come into contact. In this case the thickness of each compressed brush layer, which we denote by $d(\leq d_\mathrm{o})$, is always half the separation h, viz.,

$$h = 2d \leq 2d_\mathrm{o} \ . \qquad (25)$$

Since the density of dissociated groups increases as h decreases, it is now a function of h. We denote the density of dissociated groups after contact by $N(h)$. Following de Gennes [3], we assume that when the two brushes come into contact, they are squeezed against each other but do not interdigitate (Fig. 1b). We also assume that the brushes are contracted in such a way that the density of dissociated groups is always uniform over the compressed brush layers. That is, the product of $N(h)$ and d is constant and is always equal to $N_\mathrm{o}d_\mathrm{o}$, viz.,

$$N(h)d = N(h) \cdot \frac{h}{2} = N_\mathrm{o}d_\mathrm{o} \ . \qquad (26)$$

The potential inside the brush layer is equal to the Donnan potential with the charge density $zeN(h)$. We denote the Donnan potential in the compressed brush layer by $\psi_\mathrm{DON}(h)$, which is a function of h. Since the potential inside the compressed brush layers is constant independent of x (but depends on the separation h) (Fig. 1b), the Poisson–Boltzmann equation in the brush layer (Eq. 2) becomes

$$\frac{\mathrm{d}^2y}{\mathrm{d}x^2} = \kappa^2\left[\sinh y - \frac{zN(h)}{2vn}\right] = 0 \ , \qquad (27)$$

which is equivalent to the condition of electroneutrality in the brush layer, viz.,

$$\rho_\mathrm{el}(h) + \rho_\mathrm{fix}(h) = 0 \ , \qquad (28)$$

where $\rho_\mathrm{el}(h)$ is the charge density of fixed charges and $\rho_\mathrm{fix}(h)$ is the charge density resulting from the electrolyte ions, both being functions of h, and they are given by

$$\rho_{\mathrm{fix}}(h) = zeN(h) \tag{29}$$

$$\rho_{\mathrm{el}}(h) = -2\nu en \sinh[y_{\mathrm{DON}}(h)] \ . \tag{30}$$

In Eq. (30), $y_{\mathrm{DON}}(h) = \nu e\psi_{\mathrm{DON}}(h)/kT$ is the scaled Donnan potential in the compressed brush layer. We thus obtain from Eq. (27) (or Eq. 28)

$$\sinh[y_{\mathrm{DON}}(h)] = \frac{zN(h)}{2\nu n} = \frac{zN_{\mathrm{o}}d_{\mathrm{o}}}{\nu nh} \tag{31}$$

or equivalently

$$\begin{aligned} y_{\mathrm{DON}}(h) &= \ln\left[\frac{zN(h)}{2\nu n} + \sqrt{\left(\frac{zN(h)}{2\nu n}\right)^2 + 1}\right] \\ &= \ln\left[\frac{zN_{\mathrm{o}}d_{\mathrm{o}}}{\nu nh} + \sqrt{\left(\frac{zN_{\mathrm{o}}d_{\mathrm{o}}}{\nu nh}\right)^2 + 1}\right] \ , \end{aligned} \tag{32}$$

where Eq. (26) has been used in the last step in Eqs. (31) and (32). Note that the scaled Donnan potential $y_{\mathrm{DON}}(h)$ of the compressed brush layer is related to that of the intact brush layer by

$$\sinh[y_{\mathrm{DON}}(h)] = \sinh\left(y_{\mathrm{DON}}^{(0)}\right)\left(\frac{2d_{\mathrm{o}}}{h}\right) \ . \tag{33}$$

The interaction force $P_{\mathrm{e}}(h)$ per unit area between the two brush layers after contact may be calculated from Eq. (19) by replacing $y(h/2)$ with $y_{\mathrm{DON}}(h)$, viz.,

$$P_{\mathrm{e}}(h) = 4nkT\sinh^2[y_{\mathrm{DON}}(h)/2], \quad h \leq 2d_{\mathrm{o}} \tag{34}$$

which gives, by using Eq. (31),

$$P_{\mathrm{e}}(h) = 2nkT\left[\sqrt{1 + \left(\frac{zN_{\mathrm{o}}d_{\mathrm{o}}}{\nu nh}\right)^2} - 1\right], \quad h \leq 2d_{\mathrm{o}} \ . \tag{35}$$

Equation (35) may be rewritten in terms of the Donnan potential of the intact polymer brush layers $y_{\mathrm{DON}}^{(0)}$ with the help of Eq. (31) as

$$P_{\mathrm{e}}(h) = 2nkT\left[\sqrt{1 + \left(\sinh y_{\mathrm{DON}}^{(0)}\right)^2\left(\frac{2d_{\mathrm{o}}}{h}\right)^2} - 1\right], \quad h \leq 2d_{\mathrm{o}} \ . \tag{36}$$

Equation (35) (or Eq. 36) is the required expression for the interaction force $P_{\mathrm{e}}(h)$ per unit area between two charged brush layers after they come into contact.

The interaction force $P_{\mathrm{e}}(h)(h \leq 2d_{\mathrm{o}})$ can also be obtained by differentiating the electrostatic free energy $F(h)$ of the system with respect to the plate separation h, viz., $P_{\mathrm{e}}(h) = -\partial F(h)/\partial h$. The electrostatic free energy $F(h)$ per unit area of the plates after contact can be calculated by a method of Verwey and Overbeek [10], viz.,

$$F(h) = 2d\int_0^{\rho_{\mathrm{fix}}(h)} \psi_{\mathrm{DON}}^{(0)} \mathrm{d}\rho_{\mathrm{fix}}(h), \quad h \leq 2d_{\mathrm{o}} \ . \tag{37}$$

With the help of Eqs. (28)–(30) we finally obtain

$$\begin{aligned} F(h) = 2nkTh\Bigg(&\frac{zN_{\mathrm{o}}d_{\mathrm{o}}}{\nu nh}\ln\left[\frac{zN_{\mathrm{o}}d_{\mathrm{o}}}{\nu nh} + \sqrt{\left(\frac{zN_{\mathrm{o}}d_{\mathrm{o}}}{\nu nh}\right)^2 + 1}\right] \\ &- \sqrt{1 + \left(\frac{zN_{\mathrm{o}}d_{\mathrm{o}}}{\nu nh}\right)^2} + 1\Bigg), \quad h \leq 2d_{\mathrm{o}} \ , \end{aligned} \tag{38}$$

which gives Eq. (35). Note also that the potential energy of the electrostatic interaction between the brushes after contact is given by

$$V(h) = F(h) - F(\infty) = F(h) - F(2d_{\mathrm{o}}) + V(2d_{\mathrm{o}}), \quad h \leq 2d_{\mathrm{o}} \ . \tag{39}$$

If an approximate expression (Eq. 24) is adopted, then $V(2d_{\mathrm{o}})$ is given by

$$V(2d_{\mathrm{o}}) = \frac{64nkT}{\kappa}\tanh^2(y_{\mathrm{o}}/4) \ . \tag{40}$$

Results and discussion

In Figs. 2 and 3 we give some results of the calculation of the interaction force $P_{\mathrm{e}}(h)$ acting between two compressed brush layers as a function of reduced separation κh for several values of $zN_{\mathrm{o}}/\nu n$ at scaled brush thicknesses $\kappa d_{\mathrm{o}} = 1$ and 10. The calculation was made with the help of Eq. (35) for $h \leq 2d_{\mathrm{o}}$ and Eqs.

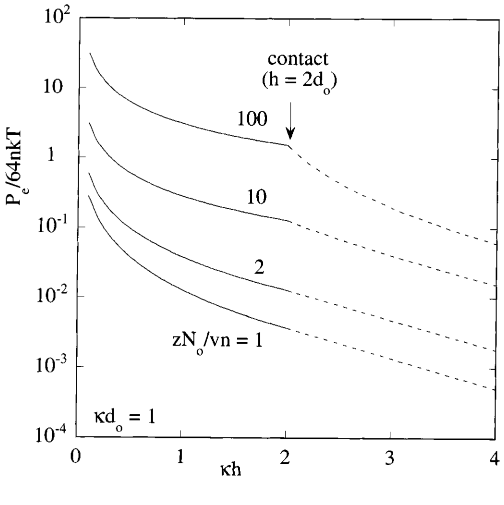


Fig. 2 Scaled repulsion $P_{\mathrm{e}}/64nkT$ per unit area between two parallel plates covered with a polymer brush layer as a function of scaled separation κh for $zN_{\mathrm{o}}/\nu n = 1, 2, 10$, and 100 (i.e., $y_{\mathrm{DON}} = 0.4812$, 0.8814, 2.312, and 4.605, respectively) at $\kappa d_{\mathrm{o}} = 1$. *Solid lines*, after contact ($h < 2d_{\mathrm{o}}$); *dotted lines*, before contact ($h \geq 2d_{\mathrm{o}}$)

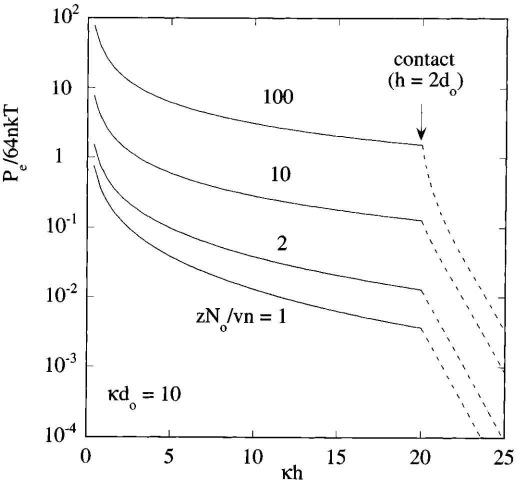


Fig. 3 Same as Fig. 2 but for $\kappa d_o = 10$

(19)–(21) for $h \geq 2d_o$. We see that the interaction force is always repulsive before and after the two brushes come into contact.

It is interesting to note that in contrast to the case of $h \geq 2d_o$, where the interaction force is essentially an exponential function of κh, the interaction force after contact of the brush layers ($h \leq 2d_o$) is a function of the inverse power of h. In order to see this more clearly we consider the following limiting cases.

1. For highly charge brushes, i.e., $|y_{\mathrm{DON}}^{(0)}| \gg 1$ or $|zN_o/vn| \gg 1$, Eq. (35) becomes

$$P_e(h) = 2nkT \left|\sinh\left(y_{\mathrm{DON}}^{(0)}\right)\right| \left(\frac{2d_o}{h}\right) = \frac{2|z|N_o d_o kT}{vh} \ . \tag{41}$$

That is, the interaction force is proportional to $1/h$. It must be stressed that the interaction force in this case becomes independent of the electrolyte concentration n.

2. For weakly charge brushes, i.e., $|y_{\mathrm{DON}}^{(0)}| \ll 1$ or $|zN_o/vn| \ll 1$, on the other hand, Eq. (35) becomes

$$P_e(h) = nkT\left(y_{\mathrm{DON}}^{(0)}\right)^2 \left(\frac{2d_o}{h}\right)^2 = \frac{(zN_o d_o)^2 kT}{v^2 n h^2} \ , \tag{42}$$

which is proportional to $1/h^2$ and to $1/n$.

3. In the limit of small separations $h \rightarrow 0$, we again have Eq. (41), viz.,

$$P_e(h) \rightarrow \frac{2|z|N_o d_o kT}{vh}, \quad h \rightarrow 0 \tag{43}$$

If, further, the total charge σ per unit area, viz.,

$$\sigma = zeN_o d_o = zeNd \tag{44}$$

(which is independent of h) is introduced, we have

$$P_e(h) \rightarrow \frac{2|\sigma|kT}{veh}, \quad h \rightarrow 0 \ . \tag{45}$$

This result agrees with the limiting behavior of the interaction force per unit area between two parallel identical plates with constant surface charge density σ [4, 11].

According to de Gennes' theory [3], the repulsive force $P_s(h)$ per unit area between two uncharged brushes of intact thickness d_o at separation h is given by

$$P_s(h) = \frac{kT}{s^3}\left[\left(\frac{2d_o}{h}\right)^{9/4} - \left(\frac{h}{2d_o}\right)^{3/4}\right], \quad h \leq 2d_o \ , \tag{46}$$

where each grafted polymer is considered as a chain of blobs of size s. Note that Eq. (46) is correct to within a numerical prefactor of order unity. The first term on the right-hand side of Eq. (46) comes from the osmotic repulsion between the brushes and the second term comes from the elastic energy of the chains. When brushes are charged, it is expected that the steric force $P_s(h)$ and the electrostatic force $P_e(h)$ both act between the charged brush layers under compression. That is, the total repulsive force $P(h)$ per unit area between the charged compressed brush layers is given by the sum of $P_e(h)$ and $P_s(h)$, viz.,

$$P(h) = P_e(h) + P_s(h) \ . \tag{47}$$

Now we compare the magnitudes of $P_e(h)$ and $P_s(h)$ with the help of Eqs. (35) and (46). We can expect that N_o and $1/s^3$ are of the same order of magnitude. Indeed, if each blob in the polymer chain has one elementary electric charge, then we have $N_o = 1/s^3$ (with $|z| = 1$). It can be shown that for highly charged brushes with $|zN_o/vn| \gg 1$, $P_e(h)$ can be comparable in magnitude to $P_s(h)$, while otherwise $P_e(h)$ has a relatively small contribution. The condition $|zN_o/vn| \gg 1$ is fulfilled for most practical cases. As a typical example, we may put $s \approx 1$ nm and

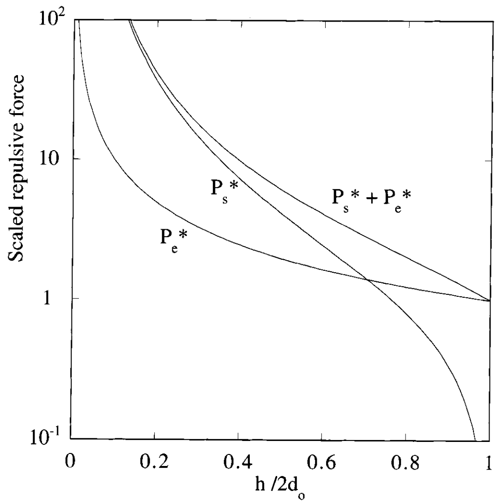


Fig. 4 Scaled repulsive force per unit area between two parallel plates covered with compressed polymer brush layers after they come into contact. $P_e^* = P_e/N_o kT$ is the scaled electrical repulsion, $P_s^* = s^3 P_s/kT$ is the scaled steric repulsion, and their sum is $P_e^* + P_s^*$

thus $N_o \approx 1/s^3 \approx 10^{27}\,\mathrm{m}^{-3}$. Thus, if, for instance, $n = 10^{-2}\,\mathrm{M} = 6 \times 10^{24}\,\mathrm{m}^{-3}$, we have $N/n \approx 1.7 \times 10^2$, and so $P_e(h)$ may be approximated by Eq. (41), which is independent of the electrolyte concentration n. In Fig. 4 we compare scaled repulsive forces $P_s{}^*(h)$ and $P_e{}^*(h)$ and their sum $P_s{}^*(h) + P_e{}^*(h)$ as a function of $h/2d_o$ for $|z| = 1$ and $v = 1$, where $P_e{}^*(h) = P_e/N_okT$ and $P_s{}^*(h) = s^3P_s/kT$. We see that P_e can be comparable in magnitude to P_s and can even exceed P_s, especially when the brushes are weakly compressed.

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