The stability of a colloidal suspension of coated magnetic particles in an aqueous solution

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Expressions for the magnetic, electrostatic and van der Waals interactions between isolated magnetic spheres which are coated with an inert material and immersed in an aqueous electrolyte solution are obtained and used to study the stability of a colloid of spheres in an electrolyte. Use is made of a simplified version of the theory of colloid stability of Derjaguin, Landau, Verwey, and Overbeek. We find that the colloidal dispersion becomes more stable as 1) the electrolyte concentration is decreased, 2) the radius of the magnetic spheres is decreased, or 3) the thickness of the inert layer is increased. In order to obtain stability with uncoated spheres, the spheres should have radii of about 5 nm. Such radii are typical of ferrofluids.

1. Introduction

A considerable advance in the understanding of colloidal systems was achieved with the Derjaguin-Landau-Verwey-Overbeek (DLVO) theory of the interaction of two colloidal particles [1]. The interaction energy was depicted as consisting of two components: an attractive London-van der Waals energy, W_{vdW}, arising from the induced dipole-dipole interactions between neutral particles, and a repulsive interaction, W_{El}, arising from the overlapping of the electric double layers in the intervening electrolyte. Assuming that these two components are independent, the total interaction free energy, V, could be expressed as

$$V = V_{\mathrm{El}} + V_{\mathrm{vdW}} \,. \tag{1}$$

For the specific case of magnetic colloids, we have extended the DLVO theory of colloidal stability to include an additive attractive magnetic dipole-dipole interaction, V_{Mag}; viz., in the presence of induced or permanent magnetic dipoles, the total interaction free energy of the two particles is assumed to be

$$V = V_{\mathrm{El}} + V_{\mathrm{vdW}} + V_{\mathrm{Mag}} \,. \tag{2}$$

Two criteria of the DLVO theory for stability are that the potential as a function of the distance of separation of the two particles should have a maximum which is considerably greater than kT and should have no potential minima deeper than kT, thus preventing agglomeration of particles. The latter ensures that kinetic energy due to thermal motion of the particles is large compared with the depth of the secondary minimum.

In our approach, the particles are encapsulated within a nonmagnetic incompressible shell. This inert layer functions

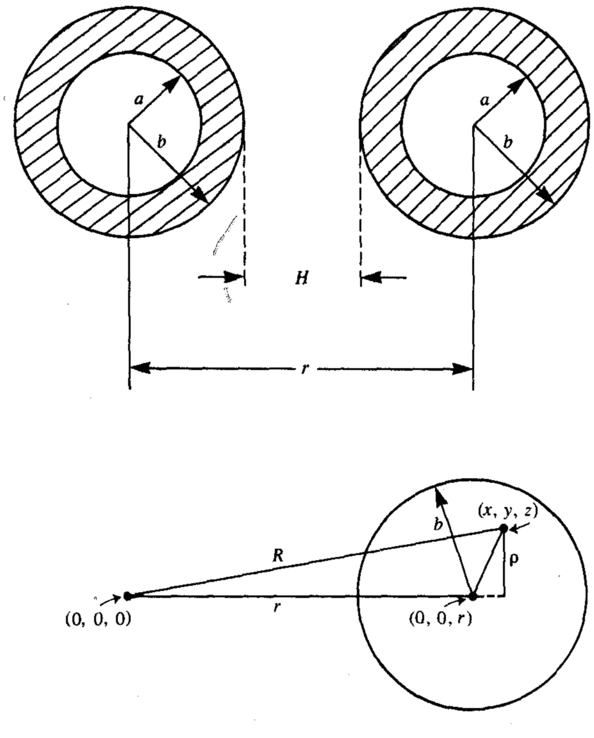

Figure 1

Geometry used in the integration to obtain I_b.

as a steric barrier which reduces the magnetic attraction between the particles and results in a considerable improvement in the stability of the dispersion. Our objective in this paper is to incorporate the magnetic dipole-dipole interaction into the DLVO theory and assess its effect upon the stability of magnetic particles. We examine the interaction free energy for two charged magnetic particles as a function of 1) salt concentration, 2) particle size, and 3) thickness of the inert coating.

2. Calculation of the interaction between a pair of colloidal particles

We consider the colloidal suspension to consist of spheres of radius b in a monovalent aqueous electrolyte solution. The ions are approximated as point charges whose charge is $\pm e$ and the solvent is approximated as a uniform dielectric medium whose dielectric constant $\varepsilon = 78.5$. The spheres are considered to be spheres of Fe_2O_3 of radius a coated with a uniform layer of an inert material of thickness $b - a$, and their surfaces are taken to be separated by a distance H (see **Figure 1**). We assume that the concentration of colloidal particles is sufficiently dilute that we need to consider only the interaction between pairs of colloidal particles.

- *The magnetic interaction*

Because of the spherical shape of the magnetic particles, we may consider them to be point dipoles of magnetic moment $\mu = 4\pi a^3 m/3$, where m is the magnetic moment per unit volume of Fe_2O_3. Thus, for a given orientation μ_1, μ_2 of the dipoles, the pair interaction is

$$u(\mathbf{r}) = -\frac{\mu^2}{r^3} D(1, 2), \tag{3}$$

where

$$D(1, 2) = 3(\hat{\mu}_1 \cdot \hat{\mathbf{r}})(\hat{\mu}_2 \cdot \hat{\mathbf{r}}) - \hat{\mu}_1 \cdot \hat{\mu}_2 \tag{4}$$

and $\mathbf{r}$ is the distance beteen the dipoles. The caret indicates that the vectors are unit vectors.

The effective magnetic interaction is obtained by averaging $u(\mathbf{r})$ over all orientations of the colloidal particles. Thus,

$$\beta V_{\text{Mag}}(\mathbf{r}) = -\ln \langle e^{-\beta u(\mathbf{r})} \rangle, \tag{5}$$

where $\beta = 1/kT$ (k is Boltzmann's constant and T is the temperature),

$$\langle e^{-\beta u(\mathbf{r})} \rangle = \frac{1}{16\pi^2} \int \exp\{\beta\mu^2 D(1, 2)/r^3\} \times \sin\theta_1 d\theta_1 d\phi_1 \sin\theta_2 d\theta_2 d\phi_2 . \tag{6}$$

Choosing the coordinate system so that $\hat{\mathbf{r}} = (\sin\theta_1, 0, \cos\theta_1)$, $\hat{\mu}_1 = (0, 0, 1)$, $\hat{\mu}_2 = (\sin\theta_2 \cos\phi_2, \sin\theta_2 \sin\phi_2, \cos\theta_2)$, we have [2, 3]

$$\begin{aligned} D(1, 2) &= 3\cos^2\theta_1 \cos\theta_2 \\ &\quad + 3\sin\theta_1 \cos\theta_1 \sin\theta_2 \cos\phi_2 - \cos\theta_2 \\ &= \frac{1}{2}\cos\theta_2 + \frac{3}{2}\cos\alpha, \end{aligned} \tag{7}$$

where

$$\cos\alpha = \cos 2\theta_1 \sin\theta_2 + \sin 2\theta_1 \sin\theta_2 \cos\phi_2 . \tag{8}$$

Hence,

$$\begin{aligned} e^{\beta u(\mathbf{r})} &= e^{xD(1,2)} = e^{x\cos\theta_2/2} e^{3x\cos\alpha/2} \\ &= \sum_{n=1}^{\infty} (2n + 1) i_n\left(\frac{x}{2}\right) P_n(\cos\theta_2) \\ &\quad \times \sum_{m=0}^{\infty} (2m + 1) i_m\left(\frac{3x}{2}\right) P_m(\cos\alpha), \end{aligned} \tag{9}$$

where $x = \beta\mu^2/r^3$, i_n and P_n are the modified spherical Bessel functions and the Legendre polynomials, respectively [4]. Now [5]

$$P_n(\cos\alpha) = \sum_{\nu=-m}^{m} \frac{(m - |\nu|)!}{(m + |\nu|)!} P_m^{\nu}(\cos 2\theta_1)\, P_m^{\nu}(\cos\theta_2) e^{i\nu\phi_2}, \tag{10}$$

and so

$$\langle e^{xD(1,2)}\rangle = \frac{1}{16\pi^2}\sum_{m,n=0}^{\infty}(2n+1)(2m+1)i_n\left(\frac{x}{2}\right)i_m\left(\frac{3x}{2}\right)$$
$$\times\left[\sum_{\nu=-m}^{m}\frac{(m-|\nu|)!}{(m+|\nu|)!}\int_0^{\pi}P_m^{\nu}(\cos 2\theta_1)\sin\theta_1 d\theta_1\right.$$
$$\times\int_0^{\pi}P_n(\cos\theta_2)\,P_m^{\nu}(\cos\theta_2)\sin\theta_2 d\theta_2$$
$$\left.\times\int_0^{2\pi}d\phi_1\int_0^{2\pi}e^{i\nu\phi_2}d\phi_2\right]. \tag{11}$$

The integrals over ϕ_1 and ϕ_2 give 2π and $2\pi\delta_{\nu 0}$, respectively. The integral over θ_2 then gives $2\delta_{mn}/(2n+1)$, where δ_{mn} is the Kronecker delta. Thus,

$$\langle e^{xD(1,2)}\rangle = \left[\frac{1}{2}\sum_{m,n=0}^{\infty}(2n+1)i_n\left(\frac{x}{2}\right)i_n\left(\frac{3x}{2}\right)\right.$$
$$\left.\times\int_0^{\pi}P_n(\cos 2\theta_1)\sin\theta_1 d\theta_1\right]. \tag{12}$$

Since

$$\cos 2\theta_1 = \cos\theta_1\cos\theta_1 + \sin\theta_1\sin\theta_1\cos\pi, \tag{13}$$

we may write [5]

$$P_n(\cos 2\theta_1) = \sum_{\nu=-n}^{n}\frac{(n-|\nu|)!}{(n+|\nu|)!}P_n^{\nu}(\cos\theta_1)P_n^{\nu}(\cos\theta_1)e^{i\nu\pi}. \tag{14}$$

Therefore,

$$I \equiv \langle e^{xD(1,2)}\rangle = \frac{1}{2}\sum_{n=0}^{\infty}(2n+1)i_n\left(\frac{x}{2}\right)i_n\left(\frac{3x}{2}\right)$$
$$\times\sum_{\nu=-n}^{n}\frac{(n-|\nu|)!}{(n+|\nu|)!}(-1)^{\nu}\int_0^{\pi}[P_n^{\nu}(\cos\theta_1)]^2\sin\theta_1 d\theta_1$$
$$=\sum_{n=0}^{\infty}(-1)^n i_n\left(\frac{x}{2}\right)i_n\left(\frac{3x}{2}\right), \tag{15}$$

where we have used

$$\sum_{\nu=-n}^{n}(-1)^{\nu} = (-1)^n. \tag{16}$$

Hence,

$$\beta V_{\text{Mag}}(r) = -\ln\left\{\sum_{n=0}^{\infty}(-1)^n i_n\left(\frac{\beta\mu^2}{2r^3}\right)i_n\left(\frac{3\beta\mu^2}{2r^3}\right)\right\}. \tag{17}$$

If $x = \beta\mu^2/r^3$ is small,

$$I = 1 + \frac{1}{3}x^2 + \frac{1}{25}x^4 + \frac{29}{11025}x^6 + \cdots \tag{18}$$

and

$$\beta V_{\text{Mag}}(r) = -\frac{x^2}{3} + \frac{7}{450}x^4 - \frac{163}{99225}x^6 + \cdots, \tag{19}$$

and if $x = \beta\mu^2/r^3$ is large,

$$I = \frac{e^{2x}}{3x^2}\left\{\frac{1}{2} + \frac{1}{3x} + \frac{1}{2x^2} + \cdots\right\} \tag{20}$$

and

$$\beta V_{\text{Mag}} = -2x + \ln(6x^2) - \frac{2}{3x} - \frac{7}{9x^2} + \cdots. \tag{21}$$

For practical applications, it is desirable to have a simple analytic expression for the magnetic interaction that does not involve infinite sums of Bessel functions. We have constructed an analytical interpolation formula, based on Eqs. (19) and (21), whose error is less than ±0.75% for all positive values of x. Our expression is

$$\beta W_{\text{Mag}} = S(x)\left(-\frac{x^2}{3}\,\frac{1}{1+\dfrac{7x^2}{150}}\right)$$
$$+\,[1-S(x)]\left[-2x + \ln(6x^2) - \frac{2}{3x} - \frac{7}{9x^2}\right], \tag{22}$$

where the "switching" function,

$$S(x) = \exp\left[-(\ln 2)\left(\frac{x}{2.4}\right)^8\right], \tag{23}$$

is simply a mathematical device to connect the large and small x expansions for V_{Mag}. The coefficients ln 2 and 2.4 were chosen so that $S(2.4) = 1/2$ and the power 8 was chosen to minimize the error. The value $x = 2.4$ was chosen because at this point, the errors in the large and small x expansions were approximately equal.

Recently, Scholten and Tjaden [6] proposed the following expression for the orientation-averaged interaction between magnetic particles:

$$U_{\text{av}}(r) = \frac{\langle e^{-\beta u(\mathbf{r})}u(\mathbf{r})\rangle}{\langle e^{-\beta u(\mathbf{r})}\rangle}. \tag{24}$$

In view of the relation

$$U_{\text{av}}(r) = \frac{\partial}{\partial\beta}\{\beta V_{\text{Mag}}(r)\}, \tag{25}$$

we can see that U_{av} is an average *interaction* energy, whereas V_{Mag} is an average *free* energy. The latter is the appropriate quantity for Eq. (2) since both V_{El} and V_{vdW} are also *free* energies. It is the total free energy which determines the stability of colloidal systems.

For weak interactions, $\beta\mu^2/r^3 < 1$, U_{av} and V_{Mag} differ by a factor of 2, i.e.,

$$V_{\text{Mag}}(r) \approx -\frac{\beta\mu^4}{3r^6}, \tag{26}$$

$$U_{\text{av}}(r) \approx -\frac{2\beta\mu^4}{3r^6}, \tag{27}$$

while in the strong interaction limit, $\beta\mu^2/r^3 > 1$, U_{av} and V_{Mag} become equal, viz.,

$$U_{\mathrm{av}} \simeq V_{\mathrm{Mag}} \approx -\frac{2\mu^2}{r^3}. \tag{28}$$

Our expansion and that of Scholten and Tjaden differ in another respect. They use $\beta\mu^2/r^3$ as an expansion parameter. Our expansion involves modified spherical Bessel functions and is equivalent to theirs but is computationally more convenient when $\beta\mu^2/r^3$ is large.

The electrostatic interaction

The colloidal particles carry a net charge, and since the particles are immersed in an electrolyte solution, there will on average be an excess of ions of the opposite charge around the colloidal particles which tend to screen the charges on the particles. The electrostatic potential on the surface of the particles when the spheres are far apart is taken to be ϕ_s relative to the bulk electrolyte.

The electric double layer interaction arises when the counter ion clouds around a pair of particles overlap. For identical particles, this interaction results in a repulsion between the particles which varies with particle separation.

We consider a 1:1 electrolyte in which the ions are treated as point charges in a solvent which is modeled as a dielectric continuum of dielectric constant ε. The electrostatic potential is taken to be given by the Poisson-Boltzmann equation [1], which reads

$$\nabla^2\phi^* = \kappa^2 \sinh\phi^*, \tag{29}$$

where $\phi^* = \beta e\phi$ is the dimensionless potential. The Debye screening length of the electrolyte κ^{-1} is given by

$$\kappa = (8\pi n e^2 \beta/\varepsilon)^{1/2}, \tag{30}$$

where n is the concentration (number/volume) of ions of either species in the electrolyte.

To determine V_{El}, it is necessary to solve (numerically) Eq. (29) for the two-sphere geometry. However, we employ a simple analytic approximation for V_{El} which is valid for large separations [7]:

$$V_{\mathrm{El}}(r) = \varepsilon\left(\frac{kT}{e}\right)^2 a(\bar{\phi}^*)^2\,\frac{e^{-\kappa H}}{r}, \tag{31}$$

where

$$H = r - 2a, \tag{32}$$

$$\bar{\phi}^* = \frac{8\tanh(\phi_S^*/4)}{1 + \left[1 - \dfrac{2\kappa a + 1}{(\kappa a + 1)^2}\tanh^2(\phi_S^*/4)\right]^{1/2}}, \tag{33}$$

and $\phi_S^* = \beta e\phi_S$ is the dimensionless surface potential. Equation (31) is strictly only valid for $\kappa H \gtrsim 1$. For $\kappa H < 1$, it overestimates the electric double layer repulsion by about 10–15%, which does not affect our results appreciably.

The van der Waals interaction

We now obtain the van der Waals interaction between two composite spheres which consist of an inner sphere of radius a coated with a layer of a different material of thickness $b - a$ using a Hamaker-type approach. If the van der Waals interaction between two phases (one of radius a_i and material i and the other of radius a_j and material j) is assumed to be

$$V_{ij} = -A_{ij}f(a_i, r, a_j), \tag{34}$$

where A_{ij} is the Hamaker constant for the i-j interaction, the van der Waals interaction between the two composite spheres is, by superposition,

$$\begin{aligned} V_{\mathrm{vdW}} = &-[A_{11}f(b, r, b) - A_{11}f(b, r, a) + A_{12}f(b, r, a)] \\ &+[A_{11}f(a, r, b) - A_{11}f(a, r, a) + A_{12}f(a, r, a)] \\ &-[A_{21}f(a, r, b) - A_{21}f(a, r, a) + A_{22}f(a, r, a)], \end{aligned} \tag{35}$$

where the subscripts 1 and 2 in A_{ij} denote the material in the outer layer and inner sphere, respectively. Gathering like terms, we obtain

$$V_{\mathrm{vdW}} = -A_{\mathrm{bb}}f(b, r, b) + 2A_{\mathrm{ab}}f(a, r, b) - A_{\mathrm{aa}}f(a, r, a), \tag{36}$$

where

$$\begin{aligned} A_{\mathrm{bb}} &= A_{11} \\ A_{\mathrm{ab}} &= A_{11} - A_{12} \\ A_{\mathrm{aa}} &= A_{11} + A_{22} - 2A_{12}. \end{aligned} \tag{37}$$

The function $f(a, r, b)$ is given by

$$\begin{aligned} f(a, r, b) &= \frac{1}{\pi^2}\int R^{-6}\,dV_{\mathrm{a}}\,dV_{\mathrm{b}} \\ &= \frac{1}{\pi^2}\int I_{\mathrm{b}}\,dV_{\mathrm{a}}, \end{aligned} \tag{38}$$

where

$$I_{\mathrm{b}} = \int R^{-6}\,dV_{\mathrm{b}}. \tag{39}$$

Referring to Fig. 1, we see that

$$\begin{aligned} I_{\mathrm{b}} &= 2\pi\int_{r-b}^{r+b} dz \int_0^{\sqrt{b^2-(z-r)^2}} \rho(\rho^2 + z^2)^{-3}\,d\rho \\ &= \frac{\pi}{2}\int_{r-b}^{r+b} [z^{-4} - (b^2 - r^2 + 2rz)^{-2}]\,dz \\ &= \frac{4\pi}{3}\,b^3(r^2 - b^2)^{-3}. \end{aligned} \tag{40}$$

Thus,

$$
\begin{aligned}
f(a, r, b) &= \frac{2}{\pi} \int_{r-a}^{r+a} dz \int_{0}^{\sqrt{a^2-(z-r)^2}} \rho I_b d\rho \\
&= \frac{8}{3} b^3 \int_{r-a}^{r+a} dz \\
&\quad \times \int_{0}^{\sqrt{a^2-(z-r)^2}} \rho(\rho^2 + z^2 - b^2)^{-3} d\rho \\
&= \frac{2}{3} b^3 \int_{r-a}^{r+a} \{(z^2 - b^2)^{-2} \\
&\quad - (a^2 - b^2 - r^2 + 2rz)^{-2}\} dz \\
&= \frac{1}{3} \left\{ b\left[\frac{a+b}{r^2 - (a+b)^2} + \frac{a-b}{r^2 - (a-b)^2}\right] \right. \\
&\quad + \frac{1}{2} \ln \left[\frac{r^2 - (a+b)^2}{r^2 - (a-b)^2}\right] \\
&\quad \left. - \frac{b^3}{r}\left[\frac{1}{(r-a)^2 - b^2} - \frac{1}{(r+a)^2 - b^2}\right]\right\}. \qquad (41)
\end{aligned}
$$

Equation (41) can be shown to be symmetric in a and b.

For most values of $H = r - 2b$, the van der Waals energy is small compared to the other interactions. However, it is large at small separations, since when $H \ll a, b, r$,

$$
f(a, r, b) = \frac{ab}{6(a+b)} \frac{1}{H}. \qquad (42)
$$

We evaluate V_{Mag}, V_{El}, and V_{vdW} using Eqs. (17), (31)–(33), (36), and (41). For Fe_2O_3, we use $m = 110$ S.I. (350 emu/cm^3). We have chosen the surface potential ϕ_S of two coated Fe_2O_3 particles at infinite separation to be 50 mV. To obtain the Hamaker constants, A_{aa}, A_{ab}, A_{bb}, for coated Fe_2O_3 in water, we use the Lifshitz [8] expression for the nonretarded van der Waals interaction. Typical values which we obtain are

$$
\begin{aligned}
A_{aa} &= Fe_2O_3 \,|\, \text{inert layer} \,|\, Fe_2O_3 \\
&= 3.73 \times 10^{-20} \text{ J}, \qquad (43)
\end{aligned}
$$

$$
\begin{aligned}
A_{ab} &= H_2O \,|\, \text{inert layer} \,|\, Fe_2O_3 \\
&= -2.89 \times 10^{-21} \text{ J}, \qquad (44)
\end{aligned}
$$

and

$$
\begin{aligned}
A_{bb} &= \text{inert layer} \,|\, H_2O \,|\, \text{inert layer} \\
&= 7.71 \times 10^{-21} \text{ J}. \qquad (45)
\end{aligned}
$$

The values for A_{aa} and A_{ab} are those appropriate for α-Fe_2O_3 in water, rather than γ-Fe_2O_3, which is used in magnetic coatings. We were unable to find any refractive index data for γ-Fe_2O_3. We expect the values of A_{aa} and A_{ab} for γ-Fe_2O_3 in water to be similar to those given above.

In **Figure 2**, values for V/kT, where V is the total predicted interaction free energy between two isolated spherical colloid particles in this model magnetic coating, are plotted for various conditions. In Fig. 2(a) the spherical particles are assumed to be 50 nm in radius and to be coated with inert layers of various thicknesses. The colloidal suspension becomes more stable as the thickness is increased. Similarly, as is seen in Fig. 2(b), decreasing the radius of the magnetic spheres, at constant thickness of the shell, increases the stability of the suspension, as is generally the case in the DLVO theory. In Fig. 2(c), we see that lowering the concentration of electrolyte increases the stability of the suspension, as is generally the case in the DLVO theory. In fact, for the particle case considered there (20-nm radius), changing the electrolyte concentration in this model can cause the suspension to pass from stability to instability. This can be seen from the appearance of a broad primary maximum of ~5 kT in height.

As in seen in Fig. 2(d) for the smaller diameters, as is characteristic of ferrofluids, the analysis indicates that the colloid should be stable without a coating. In fact, for a 5-nm radius, the magnetic interaction is small enough that it should not contribute significantly to the total interaction. Of course, the magnetic interaction would continue to be an important contributor to other properties.

The analysis carried out in this paper provides a simple quantitative estimate of the various interactions that operate between particles in a magnetic colloidal dispersion.

In the model which was used, the three different contributions to the total interaction between coated magnetic particles have very different ranges. The magnetic interaction operates on the scale of the core, viz., as ~$(a/r)^3$. The scale of the electric double layer interaction is given by the Debye length κ^{-1}, which in turn depends on the ionic strength of the suspension medium. The van der Waals component scales as ~(b/H) and is relatively unimportant except at very small separations.

For the range of magnetic particle sizes we have considered, the magnetic interaction is strongly attractive. Although the magnetic dipole-dipole interaction, Eq. (3), can either be attractive or repulsive depending on the relative orientation of the dipoles, the orientational averaging process, Eq. (5), gives the energetically favorable (attractive) orientations heavy weighting.

The role of the nonmagnetic coating is mainly that of an inert barrier that permits the electric double layer interaction to operate at larger center-to-center particle separations.

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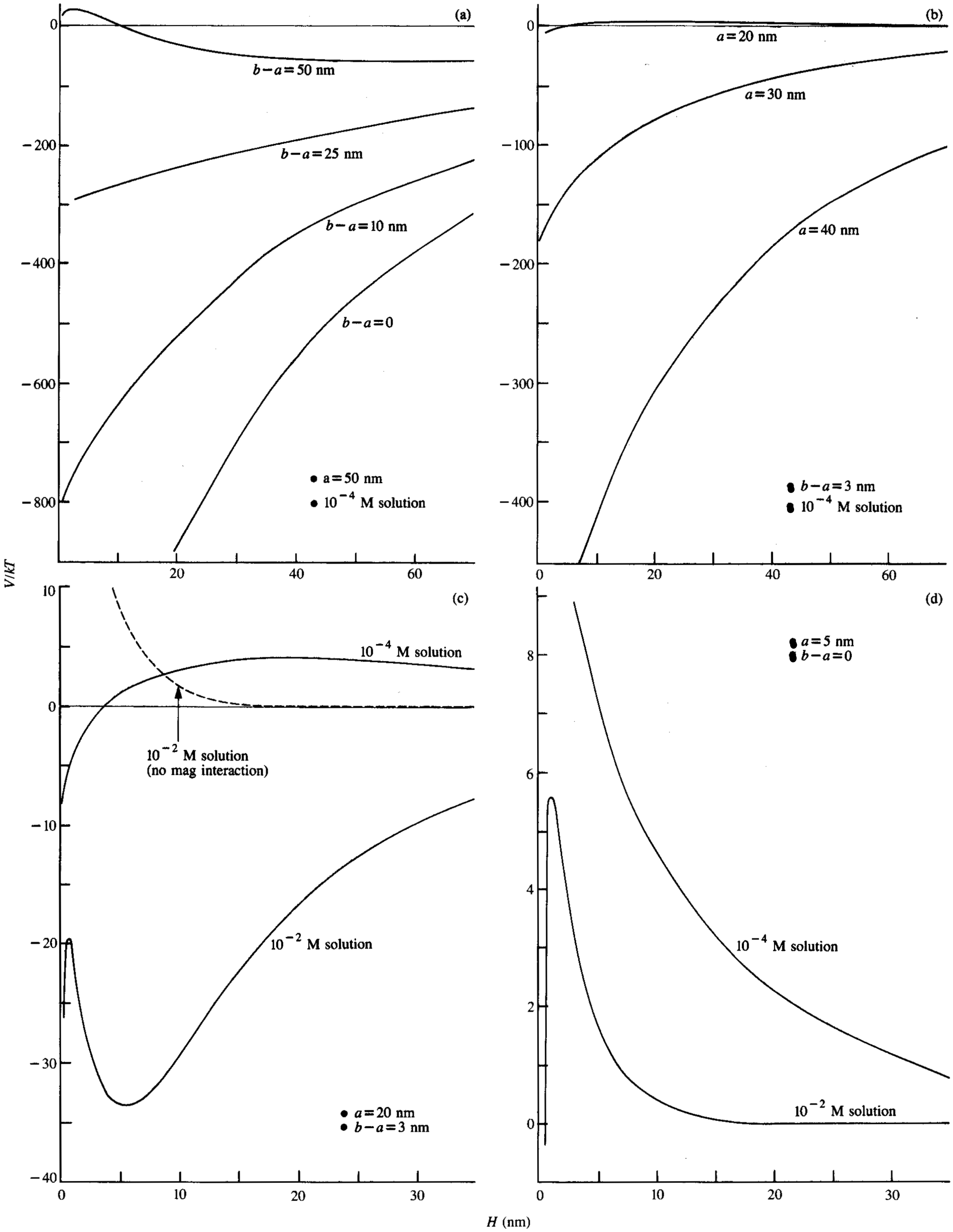

Figure 2

Predicted behavior of V/kT, where V is the total interaction free energy between two isolated γ-Fe_2O_3 spheres of the type depicted in Fig. 1 in an aqueous 1:1 electrolyte solution.

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