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Interaction free energy of small particles in an elektrolyte solution

S. Shostak[1], N. Shkoda[2], S. Stetsenko[1]

[1]Department of Electric and Electro Technology, Educational and Research Institute of Energy and Automation, National University of Life and Environmental Sciences of Ukraine, Kyiv, shostakserg@ukr.net
[2]Chuiko Institute of Surface Chemistry of the National Academy of Sciences of Ukraine, Kyiv, Ukraine

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Abstract. A solution has been constructed of the Debye-Huckel equation for system spheres with arbitrary radii and surface charges or potentials in electrolyte solutions. A general theoretical method for description of inter-particle interaction within such systems has been elaborated. The practically important case of two spheres has been considered in detail. Finite closed formulae to calculation of interaction energy of two spherical particles with constant surface charges have been obtained from general expressions within zero approximation. The known relationships of Deryagin-Landaw-Lifshits-Overbeek theory follow from our formulae in the limit cases.

Key words: Debye-Huckell equation, particles in electrolyte, pair interaction.

INTRODUCTION

Through studying ion-electrostatic interaction in systems of inorganic nanoparticles and biological cells in electrolyte solution the basic problems of calculation of the energy and forces interaction between cells and particles arise. This problem is closely connected with the problem of the electric double layer free energy of interaction between two spherical particles suspended in an aqueous electrolyte dispersion medium. From early works [1] a great attention is paid to this problem [2-7], and it is the actual question [6] until now, especially, under consideration of interaction of inorganic small particles with biological cells or microorganisms. Interaction of double diffuse layers is usually calculated on the basis of Deryaguin 's approximation. But the use of this approach can lead to incorrect results in some cases, as it was noted in [2,4].

The practical important case of two spheres is considered in detail. We fulfill the special transformation of the obtained systems. It gives the opportunity to separate the groups of connected coefficients in the infinite systems. This procedure essentially simplifies the practical solution of the problem. From general expressions the closed formulae to calculate the interaction energy of two particles with constant surface charge are received in a zero approximation. The known relations of other authors follow from our formulae as a particular case if certain conditions are fulfilled [1,2].

STATEMENT OF PROBLEM

A system of N spherical particles in an electrolyte solution with permittivity ε_m is considered. Radius of the particles is denoted as α_k, and their permittivity is denoted as ε_k $k = 1,2, \ldots, j, \ldots, N$. We link the local polar spherical coordinates $(r_k, \theta_k, \varphi_k)$ with centers of the particles (r_k is a polar radius, θ_k is an azimuth angle, φ_k is a polar angle). An arrangement of two arbitrarily chosen particles from the ensemble is shown in Figure 1, where the correspondent coordinates are indicated. Global coordinates (x, y, z) of observation point P are determined by vectors $\mathbf{r}_k$, $\mathbf{r}_j$ in the local coordinates connection, and distance between centers of the spheres $R_{kj} = |\mathbf{R}_{kj}|$, where $\mathbf{R}_{kj} = \mathbf{r}_k - \mathbf{r}_j$.

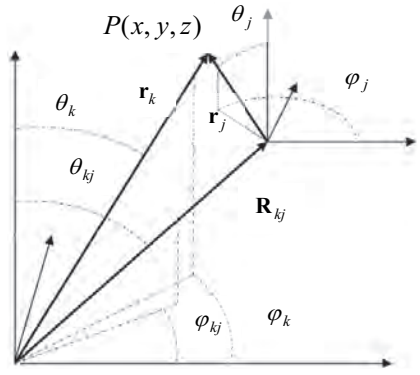

Fig. 1. The local coordinates connection

The potential corresponding to the internal and external domains of the spheres' surfaces are marked relatively by overscripts "<" and ">". In the external domain the potential $\varphi^{>}$ appears to be a sum of potentials $\phi_k^{>} = \phi_k^{>}(r_k, \theta_k, \phi_k)$, created by every sphere, i.e. $\phi^{>} = \sum_{k=1}^{N} \phi_k^{>}$ providing that any external field is absent. In the electrostatic approximation every potential $\phi_k^{>}$ ($k = 1, 2, ..., N$) is a solution of the Debye-Huckell equation (1), and potentials inside the spheres $\phi_k^{<} = \phi_k^{<}(r_k, \theta_k, \phi_k)$ the solutions of the Laplace equation (2) correspondingly:

$$\Delta\phi_k^{>} - \kappa^2 \phi_k^{>} = 0, \tag{1}$$

$$\Delta\varphi_k^{<} = 0. \tag{2}$$

Boundary conditions on the surface of the k-th sphere at $r_k = a_k$ can be formulated in different ways. We consider the case when the densities of surface charges are adjusted, so we have:

$$\phi_k^{<} = \phi^{>}, \quad \varepsilon_k \frac{\partial \varphi_k^{<}}{\partial r_k} - \varepsilon_m \frac{\partial \varphi^{>}}{\partial r_k} = 4\pi\sigma_k. \tag{3}$$

The boundary conditions (3) reflect a continuity of potentials and electric inductions on the surfaces of spheres, and densities of surface charges can be general functions of local coordinates $\sigma_k = \sigma_k(\theta_1, \phi_k)$. As usual, it is necessary to add conditions of potentials' limits:

$$\phi_k^{>} \to 0 \textbf{при } r_k^{>} \to \infty \textbf{ и } \phi_k^{<} < \infty \textbf{ при } r_k^{>} \to 0. \tag{4}$$

THE PROBLEM SOLUTION FOR SYSTEM OF N SPHERES

To solve the problem we used the expansions of solutions by series in the spherical functions $Y_{lm}(\theta_k, \varphi_k)$, $l = 0, 1, 2, ...$, $m = -l, -l+1, ..., 0, 1, 2, ..., l$. We assume that the system of the spherical functions is normalized. Inside and outside of the spheres the expansions appear to be:

$$\phi_k^{<} = \sum_{l,m} A_{lm}^{(k)} r_k^l Y_{lm}(\theta_k, \phi_k), \tag{5}$$

$$\phi_k^{>} = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} B_{lm}^{(k)} k_l(\kappa r_k) Y_{lm}(\theta_k, \phi_k). \tag{6}$$

In (6) the modified spherical Bessel functions of third kind $k_l(z)$ [10] are used.

Total potential in the surrounding media can be written as follows:

$$\phi^{>} = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} B_{lm}^{(k)} [k_l(\kappa r_k)] Y_{lm}(\theta_k, \phi_k) + \\ + \sum_{j=1}^{N}{}' \left[\sum_{l_j=0}^{\infty} \sum_{m_j=-l_j}^{l_j} B_{l_j m_j}^{(j)} [k_{l_j}(\kappa r_j)] Y_{l_j m_j}(\theta_j, \phi_j) \right], \tag{7}$$

where the stroke near the sum means that the term with subscripts $j = k$ is excluded. The sum subscripts l_j, m_j underline that they can vary independently from subscripts l, m which correspond to k -th sphere.

The problem consists in determination of unknown coefficients $A\%\backslash B™$ in the expansions of potentials (5), (6) from boundary conditions (3). To write the boundary conditions, expressions for potentials and their derivatives in other local coordinates are needed. For the solution of the problem we use the addition theorems [10]. At this, we transform the product of the spherical Bessel functions by scalar spherical functions to a product of the modified spherical Bessel functions by the scalar spherical functions. Moreover, the properties of 3-j Wigner symbols are taken into account and the transition to coefficients of Klebsh-Gordon $C^{l''m''}_{l\,m\,l'\,m'}$ [8, 9]. As result, we get the following formula for the products $k_l(\kappa r_j) Y_{lm}(\theta_j, \phi_j)$ at the condition $r_k < |\mathbf{r}_j - \mathbf{r}_k|$:

$$k_l(\kappa r_j) Y_{lm}(\theta_j, \phi_j) = \sum_{l'=0}^{\infty} , \\ \sum_{m'=-l'}^{l'} (-1)^{l'-m'} Y_{l'm'}(\theta_k, \phi_k)\, i_{l'}(\kappa r_k), \\ \sum_{l''=|l-l'|}^{l+l'} Y_{l'',m-m'}(\theta_{jk}, \phi_{jk}) k_{l''}(\kappa R_{jk}) \phi^{l\,l'\,l''}_{m\,-m'\,m-m'}, \tag{8}$$

where:

$$\varphi^{l\,l'\,l''}_{m\,-m'\,m-m'} = \left[4\pi(2l+1)(2l'+1)/(2l''+1)\right]^{1/2} \cdot \\ C^{l''\,0}_{l\,0\,l'\,0} C^{l''\,m-m'}_{l\,m\,l'\,-m'}, \tag{9}$$

and θ_{jk}, ϕ_{jk}, $\mathbf{R}_{jk}$ are shown in Fig. l. It is seen that the modified spherical Bessel function of the first kind $i_l(z)$ [10] appeared in our expression (8).

Now we can write the expression for total potential with using addition theorem (8) in the local coordinates linked with the k -th sphere:

$$\phi^{>} = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} B_{lm}^{(k)} k_l(\kappa r_k) Y_{lm}(\theta_k, \phi_k) + \\ + \sum_{j=1}^{N}{}' \sum_{l_j=0}^{\infty} \sum_{m_j=-l_j}^{l_j} B_{l_j m_j}^{(j)} \sum_{l_j'=0}^{\infty} \cdot \\ \cdot \sum_{m_j'=-l_j'}^{l_j'} (-1)^{l_j'-m_j'} Y_{l_j' m_j'}(\theta_k, \phi_k)\, i_{l_j'}(\kappa r_k) \cdot \\ \cdot \left\{ \sum_{l_j''} Y_{l_j'',m_j-m_j'}(\theta_{jk}, \phi_{jk}) k_{l_j''}(\kappa R_{jk}) \phi^{l_j\,l_j'\,l_j''}_{m_j\,-m_j'\,m_j-m_j'} \right\}, \tag{10}$$

and because the variables are separated we can found derivatives by the radial coordinates directly from (10), and then calculate their value on the surface of the k-th sphere. The expansions of the potentials and their derivatives we substitute to the boundary conditions (3). As

a result, we have the system of *2N* functional equations. Then we multiply the obtained equations by the complex conjugate functions $Y^*_{lm}(\theta_k,\varphi_k)$ and integrate over the surface of the sphere. The procedure leads to a family of infinite systems of algebraic linear equations. As the spherical harmonics are orthogonal functions, and we assume that they are normalized, we have the following values for the integrals (δ_{ij} is the Kronnecker symbol):

$$\int_0^{2\pi} d\phi \int_0^{\pi} Y_{lm}{}^*(\theta,\phi) Y_{l'm'}(\theta,\phi)\sin\theta d\theta = \delta_{ll'}\delta_{mm'}. \quad (11)$$

So, in summation by indexes l_j', m_j' the terms with subscripts $l_j' = l$, $m_j' = m$ are left only, and we get the following systems:

$$a_k^l A_{lm}^{(k)} = B_{lm}^{(k)} k_l(\kappa a_k) + $$
$$+(-1)^{l-m} i_l(\kappa a_k) \sum_{j=1}^{N}{}' \sum_{l_j=0}^{\infty} \sum_{m_j=-l_j}^{l_j} B_{l_j m_j}^{(j)}$$
$$\left\{ \sum_{l_j^*} Y_{l_j^*, m_j - m_j'}(\theta_{jk},\phi_{jk}) k_{l''}(\kappa R_{jk}) \phi^{l_j\, l_j'\, l_j^*}_{m_j\, -m_j'\, m_j - m_j'} \right\} = f_{lm}^{(k)}, \quad (12)$$

$$B_{lm}^{(k)} + \alpha_{lm}^{(k)} (-1)^{l-m} \sum_{j=1}^{N}{}' \sum_{l_j=0}^{\infty} \sum_{m_j=-l_j}^{l_j} B_{l_j m_j}^{(j)} \cdot$$
$$\cdot \left\{ \sum_{l_j''} Y_{l_j'', m_j - m_j'}(\theta_{jk},\varphi_{jk}) k_{l_j''}(\kappa R_{jk}) \varphi^{l_j\, l_j'\, l_j''}_{m_j\, -m_j'\, m_j - m_j'} \right\}, \quad (13)$$

where: $\sigma_{lm}^{(k)}$ are the expansions' coefficients of the surface charges in the spherical functions, and the notations are introduced

$$\alpha_{lm}^{(k)} = \frac{\varepsilon_k l i_l(\kappa a_k) - \varepsilon_m \kappa a_k i'_l(\kappa a_k)}{\varepsilon_k l k_l(\kappa a_k) - \varepsilon_m \kappa a_k k'_l(\kappa a_k)},$$
$$f_{lm}^{(k)} = \frac{4\pi a_k \sigma_{lm}^{(k)}}{\varepsilon_k l k_l(\kappa a_k) - \varepsilon_m \kappa a_k k'_l(\kappa a_k)}. \quad (14)$$

The primes mean a differentiation of functions by their arguments. It should be noted that for the constant densities of surface charges we have $\sigma_{lm}^{(k)} = \sqrt{4\pi}\sigma_k \delta_{l0}\delta_{m0}$.

As result, we have the aggregate of *N* connected infinite systems of linear algebraic equations. The systems contain only the coefficients $B_{lm}^{(k)}$ of external potentials and they completely solve the problem of N spheres interaction.

ION-ELECTROSTATIC ENERGY OF INTERACTION OF TWO PARTICLES

Now we consider the problem of interaction of two spheres in detail on the basis of general relationships. The line crossing the centers of spheres is taken as the axe z. The shortest distance between spheres we denote as *H,* and so the distance between centers of spheres is $d = H + a_1 + a_2$. For the Debye-Huckell approximation to the double layer free energy $F = F_{ij}$ of a pair interaction of the *i*-th and the *j*-th spheres for the known densities of the surface charges can be found using formula [3, 4]:

$$F = \frac{1}{2}\left[\int_{s_i} \sigma_i(P_i)\phi_i(P_i) dS_i + \int_{s_j} \sigma_j(P_j)\phi_i(P_i) dS_j\right],$$
$$(i, j = 1,2;\ i \neq j). \quad (15)$$

The potential energy *V* of the double layer interaction is given by the equality $V = F - F_0$ [5], where F_0 is the free energy for two single spheres, and if the densities σ_1 and σ_2 are constants:

$$F_0 = \frac{8\pi^2 a_1^3 \sigma_1^2}{\varepsilon_m(1+\kappa a_1)} + \frac{8\pi^2 a_2^3 \sigma_2^2}{\varepsilon_m(1+\kappa a_2)}. \quad (16)$$

Integrating in (15) is executed over the surface of the correspondent sphere, and because the potentials on the surfaces of the spheres are equal inside and outside the spheres, we can use either of potentials' representations. If the surface charges are constant, integration leads to calculations of the spherical functions integrals over the total surfaces of the spheres. Then we have:

$$\int_{S_j} Y_{lm} dS_j = (a_j^2 \sqrt{4\pi})\delta_{l0}\delta_{m0}, \quad (17)$$

and after integrating (16), taking into account the expansions (5), we have the simple expression for the free energy:

$$F = \sqrt{\pi}[\sigma_i a_i^2 A_{00}^{(i)} + \sigma_j a_j^2 A_{00}^{(j)}]. \quad (18)$$

So, to find the energy of pair interaction, only the first coefficients of series expansions $A_{00}^{(k)}$, $B_{00}^{(k)}$ are needed, but, as the matter of fact, their value is to be determined from the indefinite systems.

In this case, the problem is axis-symmetrical, and the system for determination of the potentials' coefficients looks like:

$$A_{00}^{(i)} = B_{00}^{(i)} k_0(\kappa a_i) + i_0(\kappa a_i) \quad B$$
$$\sum_{l'} B_{l'0}^{(j)} (-1)^{l'} (2l'+1)^{1/2} k_{l'}(\kappa d), \quad i$$
$$i, j = 1,2;\ i \neq j, \quad (19)$$

$$B_{00}^{(i)} + \alpha_0^{(i)} \sum_{l'} B_{l'0}^{(j)} (2l'+1)^{1/2} k_{l'}(\kappa d) = f_0^{(i)}, \quad (20)$$

$$B_{l0}^{(i)} + \alpha_l^{(i)} (-1)^l (2l+1)^{1/2} \sum_{l',l''} B_{l'0}^{(j)} (2l'+1)^{1/2} (-1)^{l''} k_{l'}(\kappa \quad {}^{l} \quad =$$
$$k_{l''}(\kappa d)\left(C_{l'0\,l0}^{l''0}\right)^2 = 0. \quad (21)$$

We take into account that the spheres are placed on the axe *z*, and in (12), (13) the functions $Y_{lm}(\theta,\phi)$ at $\theta = \pi$ have the value $Y_{lm}(\pi,\varphi) = \delta_{m0}(-1)^l \sqrt{(2l+1)/(4\pi)}$ [11].

The obtained system allows further simplification, because there is an opportunity to separate the coefficients $B_{l0}^{(k)}$, $B_{l0}^{(j)}$ and to get independent systems for every sphere. For all that, only the right parts of the systems define the connection between spheres.

ZERO APPROXIMATION FOR TWO SPHERES

The simplest case takes place if we keep only one term in the expansions of potentials, i.e. if $l' = m' = 0$ (a zero approximation). If the terms of higher order in comparison with the quantity $k_0^2(\kappa d)$ are neglected, the formula for the potential energy of interaction $V(d)$ can be derived:

$$V(H) = F - F_0 = \frac{8\pi^2 a_1^3}{1+\kappa a_1}\frac{\sigma_1\sigma_2}{\varepsilon_m}\cdot$$
$$\cdot\frac{k_0(\kappa d)}{z_1^2 k_0(\kappa a_1)k_1(\kappa a_2)}\left[(\kappa a_2)^2 i_0(\kappa a_2)k_1(\kappa a_2)+(\kappa a_1)^2 i_1(\kappa a_1)k_0(\kappa a_1)\right]+$$
$$+\frac{8\pi^2 a_2^3}{1+\kappa a_2}\frac{\sigma_1\sigma_2}{\varepsilon_m}\frac{k_0(\kappa d)}{(\kappa a_2)^2 k_0(\kappa a_2)k_1(\kappa a_1)}\cdot$$
$$\cdot\left[(\kappa a_1)^2 i_0(\kappa a_1)k_1(\kappa a_1)+(\kappa a_2)^2 i_1(\kappa a_2)k_0(\kappa a_2)\right]. \qquad (22)$$

In the formula (22) the modified spherical Bessel functions of the first order $k_1(z) = -k_0'(z)$, $i_1(z) = i_0'(z)$ ($k_0(z) = (\pi/2)\exp(-z)/z$, $i_0(z) = shz/z$) are used [17].

When the distances between spheres are large at $d \to \infty$ and $k_0(\kappa d) \to 0$, general formulae are simplified, and we obtain the formula (16).

Now we consider the spheres when $a_1 = a_2 = a$ and $\sigma_1 \neq \sigma_2$. At the condition $a_1 = a_2 = a$ the corresponding formula takes the form:

$$V(H) = \frac{16\pi^2 a^3\sigma_1\sigma_2}{\varepsilon_m(1+\kappa a)^2}\frac{a}{H+2a}e^{-\kappa H} +$$
$$+\frac{4\pi^2 a^3(\sigma_1^2+\sigma_2^2)}{\varepsilon_m(1+\kappa a)^3}\left[(\kappa a-1)+(\kappa a+1)e^{-2\kappa a}\right]\cdot$$
$$\cdot\left(\frac{a}{H+2a}\right)^2 e^{-2\kappa H}. \qquad (23)$$

To comparison we write out the formulae Hiroyuki Oshima [5], which had been derived as an approximation for large radii and small separation. At the condition $a_1 = a_2 = a$ the correspondent formula takes the form:

$$V_{HO}(H) = \frac{4\pi^2 a}{\varepsilon_m\kappa^2}\frac{H+a}{H+2a}\left[-(\sigma_1^2+\sigma_2^2)\ln(1-A^2)+2\sigma_1\sigma_2\ln\frac{1+A}{1-A}\right], \qquad (24)$$

where: $A = \left(a/(a+H)\right)\exp(-\kappa H)$.

If $\sigma_1 = \sigma_2 = \sigma$, it follows from (24) that:

$$V_{HO}(H) = -F_0\frac{(1+\kappa a)}{(\kappa a)^2}\cdot\frac{H+a}{H+2a}\ln\left(1-\frac{a}{H+a}e^{-\kappa H}\right), \qquad (25)$$

where: in this case $F_0 = 16\pi^2 a^3\sigma^2/[\varepsilon_m(1+\kappa a)]$.

The Derjaguin's metod [1] gives the following simple expression for the same case:

$$V_D = -F_0\frac{(1+\kappa a)}{(\kappa a)^2}\ln(1-e^{-\kappa H}). \qquad (26)$$

If we neglect the second term in our formula (23), it takes the form:

$$V(H) = F_0\frac{1}{1+\kappa a}\frac{a}{H+2a}e^{-\kappa H}. \qquad (27)$$

We see, at $\kappa H \to 0$, then $V_D \to \infty$. It means that formula (26) can give incorrect results at small κH values. The analogous situation takes place for relationship (25), too. When $\kappa H >> 1$ and $\kappa a >> 1$, after expanding the logarithms to correspondent series and keeping the first

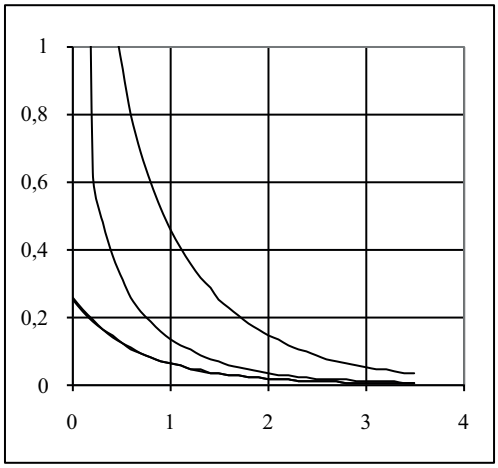

Fig. 2. Interactions' energy $V^* = V/V_0$ of two identical spheres with the constant charges versus κH at $\kappa a = 1$

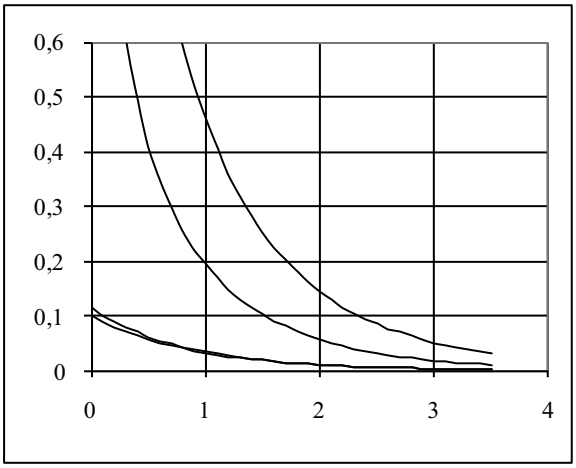

Fig. 3. Interactions' energy $V^* = V/V_0$ of two identical spheres with the constant charges versus κH at $\kappa a = 4$

1 – Derjaguin approximation; 2 – Hiroyuki Oshima approximation; 3 – our simplest zero approximation; 4 – our improved zero approximation.

terms of the series we get the same expressions from formulae (25), (27).

NUMERICAL RESULTS

We obtain closed formulae with results of another author in the limit cases, but our results are more general and they can be improved if the next approximation is taken into account. Some results of the worked out calculation are presented in Fig. 2 and Fig. 3.

We consider the case of adjusted densities of surfaces charges. We determine dependences of non-dimensional interaction energy , where $V_0 = 16\pi^2 a^3 \sigma^2 / (\varepsilon_m (1+\kappa a))$, for two identical spheres with constant and equal charges versus parameter κH at κa = 1 (Fig. 2) and at κa = 4 (Fig. 3).

It follows from this data that at $\kappa H > 3$ formulae H. Oshima (25) and our formulae for zero approximation give the neighbor values, but Derjaguin's formula gives a too high value. All the results begin to agree at large values of parameter κH. Our results strongly differ from results of Hiroyuki Oshima and Derjaguin, especially at very small values of κH, because and $V_D \to \infty$ and $V_{HO} \to \infty$, when $\kappa H \to 0$.

CONCLUSIONS

The exact solution for interaction of a system of small spherical particles in electrolyte is obtained. On the basis of the exact solutions the handily closed formulae for calculating ion-electrostatic energy of two spheres are derived. Our results correspond to the results of other authors in simple cases and generalized ones in range of small values parameter κa. In this article we have considered the case when surfaces' charges are given, but the problem of spherical particles interaction with giving surface potentials can be solved similarly.

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