

Influence of the electrolyte concentration on interaction between silica particles*

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The destruction time of the sediment column structure was measured for different fractions of silica particles in NaCl and CaCl₂ solutions whose concentrations were varied from 0.25 M to 2.0 M. A linear relationship between the reciprocal of the destruction time of the silica sediment column structure and average diameter of the silica fraction, the density and work of cohesion of the solutions was found. The determined values of the particle diameter, the cohesion work and density of the solution calculated from these linear dependences for the time equal to infinity ($1/t = 0$) were named critical. Using the above critical parameters the detachment force of one particle from another was calculated. For the system whose components had critical values the detachment force was found to be equal to the attachment force resulting from the interfacial interaction. On the basis of our earlier study and Derjaguin's approximation the radius of the contact plane and the attachment force between two silica particles were calculated. The detachment force decreased with increasing salt concentration, but the attachment force was nearly constant. It was found that for a given salt concentration the relationship of the reciprocal of the destruction time as a function of the difference between the detachment and attachment forces was also linear. It appeared that for the silica column structure having critical parameters the values of the difference between the detachment and attachment forces were negative. This means that in the calculation of the attachment force between silica particles in the studied solutions an additional interaction should be taken into account.

* Dedicated to Professor Emil Chibowski on the occasion of his 65th birthday.

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1. INTRODUCTION

Adhesional interactions between solid particles through a liquid film are determined by the surface and interface properties of the components of the studied system. The interactions are important to understand many phenomena connected with the chemistry of such processes as stability of colloids and suspension, agglomeration, adhesion, adsorption, lubrication, wettability and others.

The physicochemical properties of silica, as well as silicate minerals depend on their origin and their environmental conditions which influence the stability of their dispersion systems. Therefore, the surface and interface properties of these minerals are important in many scientific and technological areas. Changes of these properties depend not only on the size of the particles and values of the contact plane between them but also on the particular components of the studied systems, and they determine the values of interactions between silica particles. The interactions, or commonly expressed adhesion forces are treated as attractive van der Waals and repulsive forces due to a layer of structured water at the silica- water interface [1-3], and they determine the adhesion phenomena in silica-liquid-silica systems.

Different theoretical approaches [4-7] and many sophisticated techniques and apparatuses [4,5, 8-13] are used to characterize the adhesional interactions, whereas in our laboratory a very simple device was used to measure the destruction time of the mineral sediment column structure formed in water [14-15]. The time is proportional to the attachment and detachment forces between mineral grains

Using this device, we measured the destruction time of the structure of the coal and silica sediment column formed in homologous series of alkane and alcohol [16- 20]. We found that the time to be proportional to the attachment and detachment forces between mineral grains, and the detachment force of particle from one another in the studied liquids occurred by disruption the liquid film present between them, and the free energy of the film was slightly changed by the surface free energy of the studied solids.

The water dispersion system, containing among other things silica, may consist of different inorganic compounds dissolved in water. Adsorption of these substances at solid-liquid interface influences the values of adhesion between solid particles through a liquid film. The goal of the study was to estimate the influence of the concentration of inorganic salts on the interaction between silica particles through a liquid film, To calculate the interaction values, the destruction time of the silica sediment column structure was measured in NaCl and CaCl₂ solution.

2. EXPERIMENTAL

Method and materials. The device for measurement of the destruction time of the sediment column structure of particles of a solid was described in details elsewhere [14-20]. The main parts of the device are a glass tube in which the sediment column is formed and a glass vessel connected by a rubber plug. Here, we present only the way how the destruction time measurements were done.

The destruction time of the sediment column structure was measured for high purity silica powder (>99.99%), which came from naturally occurring Russian silica particles (Caucasus). The silica powder, kindly supplied by the Glassworks "Ożarów" (Poland), was fractionated by screening into four narrow fractions of average diameters: $1.04 \cdot 10^{-4}$ m, $1.35 \cdot 10^{-4}$ m, $1.62 \cdot 10^{-4}$ m and $2.25 \cdot 10^{-4}$ m. To remove the contamination from the surface occurring during screening each fraction was immersed in HCl solution at a dilution of 1:1 for 24 h, stirring the suspension from time to time, and then the particles were washed with redistilled and deionized water until pH and conductivity were the same as that of the water used for cleaning. After that the silica particles were heated for 4h at 450° C, and the dried products were stored in hermetically closed jars.

The measurements of the destruction time of the sediment silica column structure were carried out in NaCl and CaCl₂ solutions at concentrations ranging from 0.25 M to 2.0 M. The solutions of the salts were prepared in doubly distilled and deionized water from a Milli-Q system, and the pH of the solutions was natural.

Measurements. The sediment column of silica particles was formed in the glass tube by pouring $3.0 \cdot 10^{-5}$ m³ of NaCl or CaCl₂ solution at a given concentration into the glass vessel and adding $1.388 \cdot 10^{-3}$ kg sample of the studied fraction of silica particles. The particles falling in the tube formed a sediment column, and its structure was established for 10 min. Then the device was quickly inverted, and single silica particles detached from the sediment column dropped at the bottom of the tube forming a new sediment column.

The destruction time of the silica sediment column structure was measured by a stopwatch, starting when the first silica particle detached from the column, and stopping when the last particle started to drop. The destruction time measurements were repeated several times for each studied system. The reproducibility of the measurements was very good.

The surface tension of the electrolyte solutions and their density were measured by the weight–volume method and a densitometer, respectively. These measurements were carried out at 20° C ($\pm 0.5^\circ$ C), and the results obtained were nearly the same as those in the literature [21,22].

3. RESULTS AND DISCUSSION

The measured values of the destruction time of the silica sediment column structure were expressed for the sediment column formed from $1.0 \cdot 10^{-3}$ kg silica sample and they are listed in Tab. 1A and 1B for NaCl and CaCl₂ solution, respectively. Afterwards the reciprocal values of the destruction time were calculated.

Tab. 1. Values of the destruction time in [s] of the silica sediment column structure formed in NaCl (part A) and CaCl₂ (part B) solutions.

Salt Concentration M	Average diameter of the silica particle fractions ($\times 10^{-5}$ [m])							
	A				B			
0.25	10.4	13.5	16.2	22.5	10.4	13.5	16.2	22.5
0.5	76.6	53.7	38.4	22.4	79.6	53.9	32.9	22.4
0.75	82.0	57.6	40.7	24.6	92.3	57.7	35.5	23.7
1.0	84.5	58.9	42.4	26.3	112.6	68.1	43.2	27.7
2.0	87.8	60.1	46.3	26.5	120.3	73.8	45.8	30.4
	92.1	63.7	49.4	28.9	144.2	95.4	59.5	39.8

The results obtained were discussed in terms of the bulk and surface properties of the components of the studied systems, i.e. the average diameter of silica particle fractions, density and work of cohesion of the solutions. In Tab. A and 2B the measured values of the densities and the surface tension of the NaCl and CaCl₂ solutions are listed, respectively, and as mentioned the results were the same as those in the literature [21, 22].

Tab. 2. Measured values of the surface tension and density for NaCl (column A) and CaCl₂ (column B) solutions.

Concentration M	Surface tension γ mN/m		Density kg/m ³	
	A	B	A	B
0	72.4	72.4	999.2	999.2
0.25	73.3	73.4	1013.5	1020.9
0.5	74.0	74.2	1026.8	1043.1
0.75	74.5	75.4	1040.8	1064.4
1.0	75.1	76.8	1054.3	1086.9
2.0	75.9	78.7	1078.2	1167.0

The destruction time of the silica sediment column structure as a function of the average diameters of silica fractions, density and work of cohesion of solutions increased with increasing density and work of cohesion of the

electrolyte solutions and decreased with increasing average diameter of the silica fractions.

However, the trends of the reciprocal of the destruction time, ($1/t$), of the silica sediment column structure as a function of the above-mentioned parameters, were inverse on relation to those for the destruction time, but the dependences were linear as presented in Figures 1, 2 and 3. As can be seen, the reciprocal of the destruction time increases linearly with increasing average diameter of the silica fractions (Figure 1), and decreases linearly with increasing density (Figure 2), and the work of cohesion (Figure 3) of both electrolyte solutions. These facts suggest that the difference between the detachment and attachment forces should decrease with increasing density and work of cohesion of electrolyte solutions.

Determination of the critical parameters. As can be seen from Figures 1, 2 and 3, no the silica particles detach from those in the columns even for infinitely long time ($1/t = 0$). The curves presented in these figures satisfy the equation [16-20]:

$$1/t = A + B \cdot X \quad (1)$$

where: t is the destruction time, A and B are the constants and X is the independent variable (i.e. either the average diameter of the silica particle fraction (Figure 1), the density of solutions (Figure 2) or its work of cohesion (Figure 3)).

The critical diameter values ($t = \infty$) for silica particles were calculated from equation (1) using the results presented in Figure 1A and 1B for NaCl and CaCl₂ solutions, respectively. The obtained values of the critical diameter do not depend on the concentration of salts, and they are in the ranges from $6.1 \cdot 10^{-5}$ m to $5.0 \cdot 10^{-5}$ m and from $6.6 \cdot 10^{-5}$ m to $6.0 \cdot 10^{-5}$ m for NaCl and CaCl₂ solutions, respectively. The average critical diameters of the silica particles calculated for both solutions are $5.4 \cdot 10^{-5}$ m and $6.3 \cdot 10^{-5}$ m, respectively.

The values of the critical densities were calculated from equation (1) on the basis of the results presented in Figure 2A and 2B for NaCl and CaCl₂ solutions, respectively. The obtained critical densities did not depend on the average diameter of silica particles and they range from 1361 kg/m^3 to 1253 kg/m^3 , and the average value was 1309 kg/m^3 for NaCl solutions. As it is seen from Figure 2B the critical densities for CaCl₂ solutions were nearly constant, and the average value was 1344 kg/m^3 .

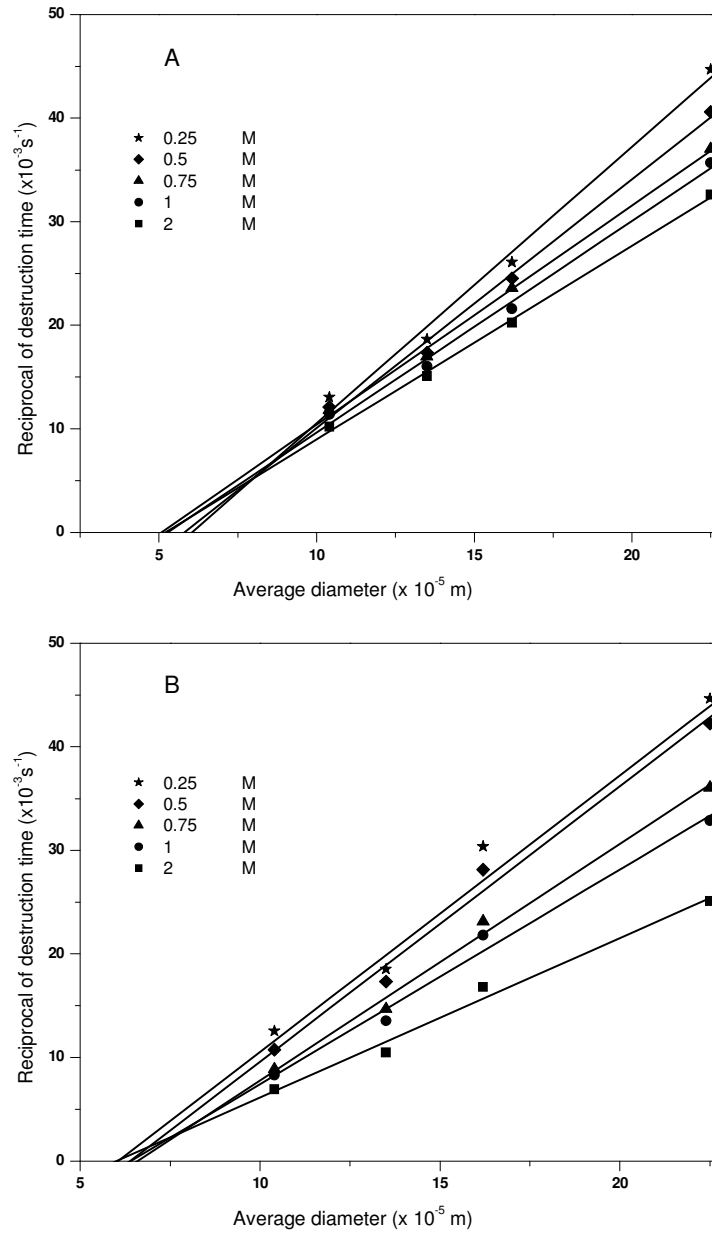


Fig. 1. Reciprocal of the destruction time of the sediment column structure ($1/t$ in $1/s$) as a function of the average diameter of silica particles. The destruction time measured in NaCl (Figure A) and CaCl₂ (Figure B) solutions.

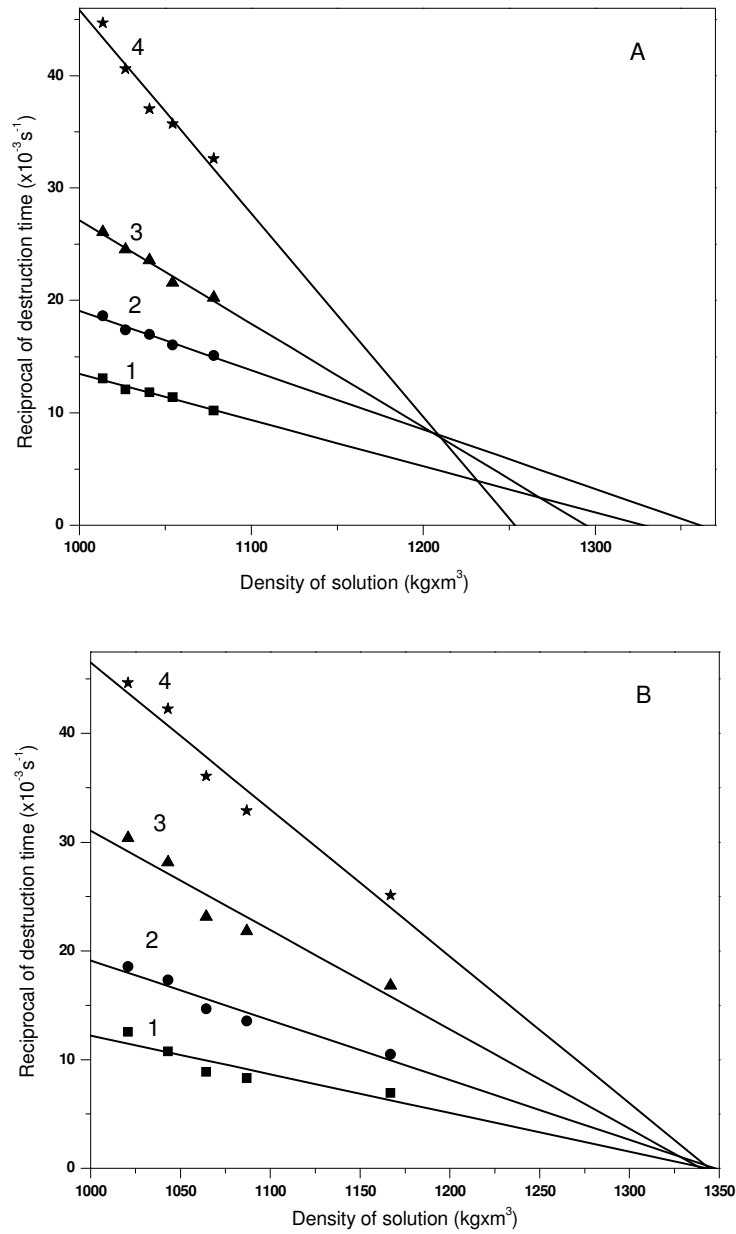


Fig. 2. Reciprocal of the destruction time of the sediment column structure ($1/t$ in $1/s$) as a function of the density of NaCl (Figure A) and $CaCl_2$ (Figure B) solutions. Curves 1, 2, 3 and 4 represent the changes in $1/t$ for silica particles having the average diameters: $1.04 \cdot 10^{-4}$ m, $1.35 \cdot 10^{-4}$ m, $1.62 \cdot 10^{-4}$ m and $2.25 \cdot 10^{-4}$ m, respectively.

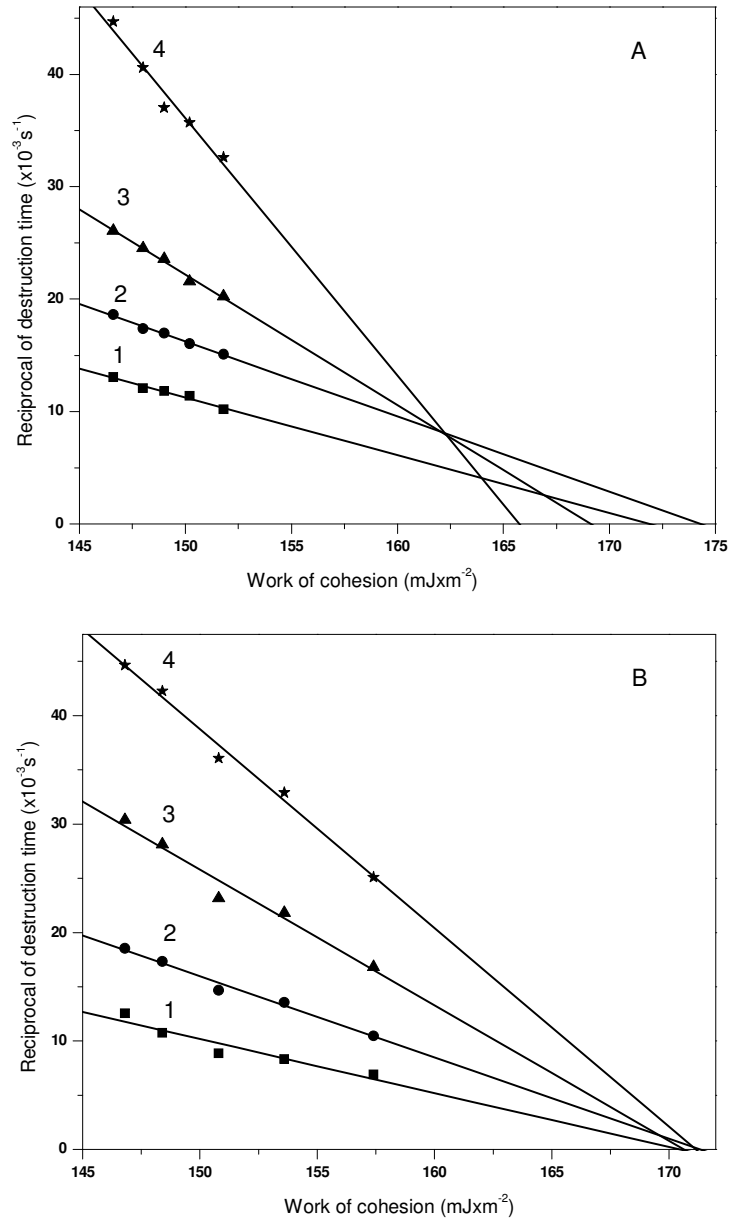


Fig. 3. Reciprocal of the destruction time ($1/t$ in $1/s$) as a function of the work of cohesion of NaCl (Figure A) and CaCl₂ (Figure B) solutions. Curves 1, 2, 3 and 4 represent the changes in $1/t$ for silica particles having the average diameters: $1.04 \cdot 10^{-4}$ m, $1.35 \cdot 10^{-4}$ m, $1.62 \cdot 10^{-4}$ m and $2.25 \cdot 10^{-4}$ m, respectively.

The critical work of cohesion calculated from equation (1), using the results presented in Figure 3A and 3B, did not depend on the average diameter of silica particles, either, and it ranged from 174.3 mJ/m² to 167.7 mJ/m² and from 171.3 mJ/m² to 170.4 mJ/m² for NaCl and CaCl₂ solution, respectively. The average values of the critical work of cohesion were nearly the same for both electrolytes and they were 170.25 mJ/m² for NaCl and 170.85 mJ/m² for CaCl₂ solutions. Although the results were unexpected, in our opinion they were reasonable taking into account the influence of an inorganic salt on the change of the surface tension of its solution. Therefore, for further calculation the average value, 170.5 mJ/m² of the critical work of cohesion was used for both electrolyte solutions.

Silica particles of a diameter $\leq d_c$, or the sediment column formed in the solution whose density or work of cohesion was critical, did not detach from those of the column, so for such particles the detachment force should be smaller or at least equal to the attachment force.

Calculation of the detachment force. Assuming a spherical shape of silica particles, their detachment force (F_D) from those forming the sediment column in the electrolyte solutions can be calculated from the equation:

$$F_D = m \cdot g - V \cdot \rho_L \cdot g = 1/6 \cdot \Pi \cdot d_c^3 g(\rho_s - \rho_L) \quad (2)$$

where: m is the silica particle mass, g is the acceleration due to gravity, V is the silica particle volume, ρ_s and ρ_L are the densities of the silica and solution, respectively, and d_c is the average critical particle diameter.

As we mentioned above, for particles having the critical diameter the detachment and attachment forces are equal, and the sediment column structure is thus stable.

The detachment force (F_D), dependent on the gravity force, was calculated from equation (2), and their values decreased with increasing NaCl and CaCl₂ concentrations from 13.2·10⁻¹⁰ N/particle to 11.2·10⁻¹⁰ N/particle and from 20.9·10⁻¹⁰ N/particle to 16.7·10⁻¹⁰ N/particle, respectively. The smallest values of the detachment forces were obtained for the solutions whose densities were critical. There were large differences between the F_D values for NaCl and CaCl₂ solutions, which directly resulted from big differences in the values of the critical diameters of silica particles obtained in these two kinds of solutions. Accordingly, the difference in the values of the critical diameter of silica particles may be caused by different enthalpies of hydration of monovalent and bivalent cations (Na^+ and Ca^{2+}) [23-29] or by different hydration of the silica surface occurring during the destruction time measurements. These suggestions

are confirmed by different values of the critical diameter of the particles of hydrophobic and hydrophilic silica [19, 20]. They may cause changes of the value of the repulsive interaction between silica particles through the film of electrolyte solution.

Calculation of the attachment force. For the sediment column formed of particles having the critical diameter the detachment force satisfies the equation:

$$F_D = W_{Coh.C} \cdot L = W_{Coh.C} \cdot 2\Pi \cdot R \quad (3)$$

where: $W_{Coh.C}$ is the critical work of cohesion of liquid, L is the perimeter of the contact plane between two solid particles and R is the radius of the contact plane.

To calculate the attachment force of one silica particle to another in the studied solutions, at first we had to know the radius of the contact plane between them. The values of the radii were calculated from equation (3) using the same value of the average critical work of cohesion for both (NaCl and CaCl₂) solutions, and they decrease from $12.4 \cdot 10^{-10}$ m to $11.9 \cdot 10^{-10}$ m, and from $19.6 \cdot 10^{-10}$ m to $17.8 \cdot 10^{-10}$ m with increasing NaCl and CaCl₂ concentration from 0.25 M to 2 M, respectively. For the NaCl and CaCl₂ solutions having the critical work of cohesion, the radii of the contact planes were $10.5 \cdot 10^{-10}$ m and $15.7 \cdot 10^{-10}$ m, respectively.

On the basis of Derjaguin's approximation [5, 8, 9] and knowing the radius of the contact plane between two particles through the liquid in the sediment column, the attachment force satisfies the equation:

$$F_A = W_{Coh.} \cdot 2\Pi \cdot R \quad (4)$$

where $W_{Coh.}$ is the work of cohesion of the solution.

In equilibrium conditions the attachment force, F_A , between two silica particles in the solutions satisfies equation (4). The values of the attachment force, F_A , calculated from this equation range from $11.4 \cdot 10^{-10}$ N/particle to $11.3 \cdot 10^{-10}$ N/particle and from $18.0 \cdot 10^{-10}$ N/particle to $17.6 \cdot 10^{-10}$ N/particle for the NaCl and CaCl₂ solutions, respectively, and as it is seen they are practically constant. The values of the attachment force, F_A , between silica particles in the solutions (NaCl and CaCl₂), whose cohesion work is critical, are equal to the smallest value of the detachment force, F_D , which were calculated for solutions having the critical density. The difference in the values of the critical forces obtained for the studied solutions is clear, i.e. $11.2 \cdot 10^{-10}$ N/particle and $16.7 \cdot 10^{-10}$ N/particle for NaCl and CaCl₂ solutions, respectively. This difference results from different values of the contact planes between the silica particles in

these solutions, which in turn were determined by the critical diameter values of the silica particles. It should be emphasized that the values of the critical work of cohesion for NaCl and CaCl₂ solutions are practically the same (170.25 mJ/m² and 170.85 mJ/m²), and the difference in their critical densities is not so large (1309.0 kg/m³ and 1344.0 kg/m³). These facts support our earlier suggestion concerning the detachment forces that such big differences in the attachment forces between silica particles in the studied solutions can also be caused by different enthalpies of hydration of Na⁺ and Ca²⁺ cations and by different hydration of the silica surface [19, 23-29]. These differences can alter the values of the attractive interaction. This suggestion should be confirmed by measurements of the influence of temperature on the destruction time of the silica column structures in the salt solutions and by electrochemical tests.

The calculated differences between the detachment and attachment forces ($\Delta F = F_D - F_A$) for NaCl and CaCl₂ solutions decreased from $1.8 \cdot 10^{-10}$ N/particle to $14 \cdot 10^{-10}$ N/particle and from $2.8 \cdot 10^{-10}$ N/particle to $14 \cdot 10^{-10}$ N/particle, respectively, as the concentrations of the salts solutions increased from 0.25 M to 2 M.

The relationship between the reciprocal of the destruction time of the silica sediment column structure as a function of the differences of the detachment and attachment forces, (ΔF), is presented in Figure 4A and 4B for NaCl and CaCl₂ solutions, respectively. As can be seen the curves in Figure 4A and 4B are linear and they satisfy equation 1. The critical values of the differences, (ΔF_C), (destruction time is equal to infinity) calculated from this equation range from $-0.2 \cdot 10^{-10}$ N/particle to $-0.6 \cdot 10^{-10}$ N/particle and from $-0.3 \cdot 10^{-10}$ N/particle to $-0.5 \cdot 10^{-10}$ N/particle for NaCl and CaCl₂ solutions, respectively. As it is seen, the ΔF_C values are negative and they do not depend on the average diameter of the silica particle fractions. Moreover, it appeared that the average ΔF_C value for both solutions was the same amounting $-0.4 \cdot 10^{-10}$ N/particle.

The negative values of the ΔF_C for the stable silica column structure ($1/t = 0$ and $F_D = F_A$) showed that the adhesion interactions in the studied systems were determined not only by inertial and van der Waals' forces, as in the case when the liquid media were alkanes or alcohols [16-20], but also from the hydration force [23-29]. This force originates from the different enthalpies of hydration for Na⁺ and Ca²⁺ cations and silica surface hydration occurred during the destruction time measurements which could alter the attractive or repulsive interactions. So, the hydration forces and the interactions should be taken into account for calculation of the detachment and attachment forces between silica particles in the studied solutions. We shall try to solve this problem in the future by a more detailed study .

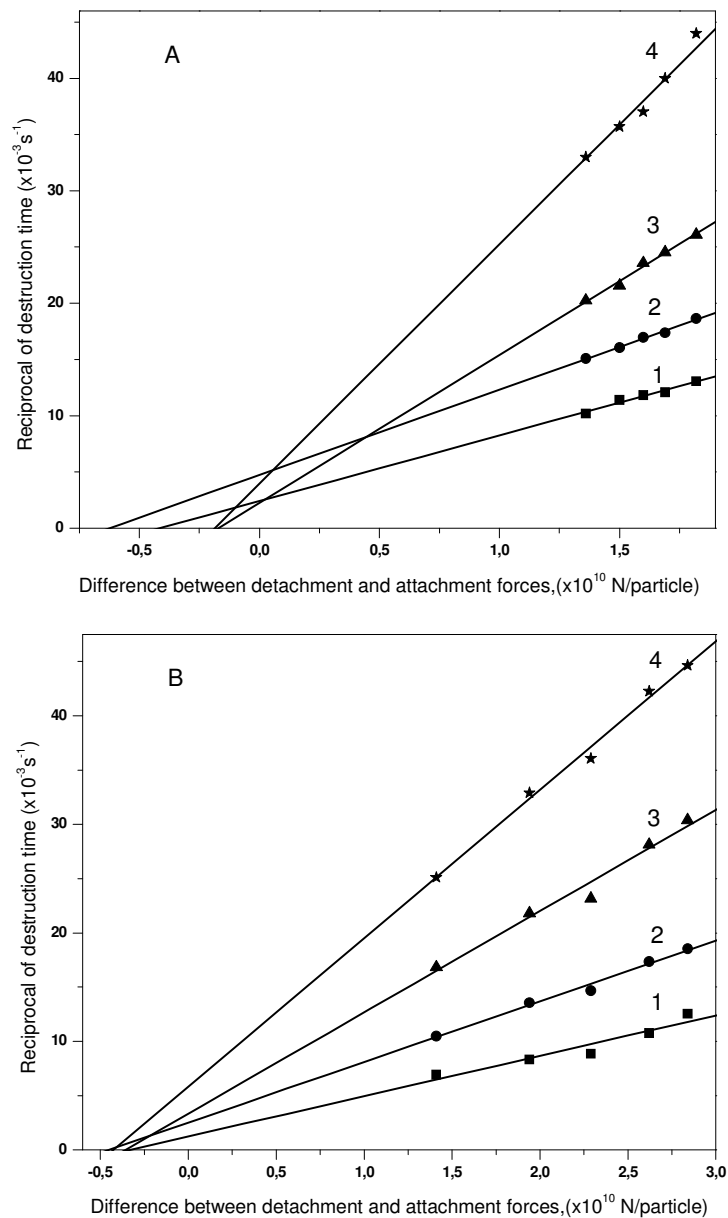


Fig. 4. Reciprocal of the destruction time ($1/t$ in $1/s$) as a function of the difference between the detachment and attachment forces (ΔF in $N/particle$) for silica particles in NaCl (Figure A) and $CaCl_2$ (Figure B) solutions. Curves 1, 2, 3 and 4 represent the changes in $1/t$ for silica particles having the average diameters: $1.04 \cdot 10^{-4}$ m, $1.35 \cdot 10^{-4}$ m, $1.62 \cdot 10^{-4}$ m and $2.25 \cdot 10^{-4}$ m, respectively.

4. CONCLUSION

On the basis of the results and discussion presented above some conclusions can be drawn. Like in the systems where the media were alkanes or alcohols, in the case of electrolyte solutions also linear dependences of the reciprocal of the destruction time on average diameters of silica fractions, the density and the work of cohesion of the solutions were found. The values of the critical parameters determined from these linear dependences, i.e. the diameter of silica particles, the density and work of cohesion of the solution enabled us to calculate the radii of the contact planes between silica particles and then the attachment forces between silica particles through the liquid film. The detachment forces of one silica particle from another in the sediment column were calculated from gravitational interaction.

If the system with one critical parameter (average diameter, density or critical work of cohesion) the detachment force is equal to the attachment force, then the structure of the sediment column is stable, and detachment of a silica particle from another occurs by disruption of the liquid film of the electrolyte solution. The value of the contact plane between silica particles depends on the kind of electrolyte, which determines the values of the detachment and attachment forces.

The negative values of the difference between the critical detachment and attachment forces, (ΔF_C), determined from the linear relationship between the reciprocal of the destruction time and the difference between the detachment and attachment forces, suggest that the hydration force resulting from different enthalpies of hydration of monovalent and bivalent cations or from hydration of the silica particles occurring during the measurements should be taken into account to calculate the attachment forces between the particles.

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