

COMPUTATIONAL APPRAISAL OF CAPILLARY PRESSURE IN LIQUID INTERLAYERS BETWEEN PARTICLES OF DISPERSIVE COMPONENTS

Yu.G. Tselishev, V.A. Valtsifer, V.N. Strelnikov

*Institute of Technical Chemistry of UB RAS,
Acad. Koroleva, 3, Perm, Russia
tselishch@yandex.ru*

A computational model comprised of two particles connected with aid of a liquid interlayer served as a basis for investigation in capillary pressure value in the liquid and in basic parameters of particles influencing this value. Both water absorbed from the environment, and liquid components of dispersive compositions can form this interlayer. Interaction between variously shaped and sized particles was investigated. The offered computational model assisted with numerical methods can be used to explore how capillary pressure and parameters of both dispersive and liquid components influence values of capillary adhesion force between particles in powder-like compositions.

Introduction

Capillary forces are the most significant forces acting between particles of dispersive components during their storage, processing, and use under natural conditions. These forces are generated in the liquid interlayer between particles that is formed, in particular, by either moisture absorbed by powders from the environment, or liquid components of dispersive compositions within a narrow gap between particles. Values of these forces are dependent on various parameters characterizing powder-like materials and the environment; at this point, capillary pressure in the liquid between particles is a parameter insufficiently investigated so far [1-3].

This work aims at investigation in how basic parameters of dispersive particles influence capillary pressure and, consequently, values of capillary forces acting in the liquid interlayer between particles of powder-like compositions.

Calculation part

Two particles with the liquid interlayer between them (Fig. 1) were opted as the computational model. At this point, the interlayer can be formed by both moisture absorbed by any powder from the environment under real conditions of storage and processing, and liquid components of dispersive composite materials.

Interaction of a liquid's molecules between themselves and dispersive particles results in the distorted meniscus of the liquid's surface

[4-8], this phenomenon causing additional pressure (P) that, in accord with the Laplace equation, equals:

$$P = p_l - p_g = \sigma \left(\frac{1}{r_1} + \frac{1}{r_2} \right), \quad (1)$$

here, p_l, p_g – pressure values in the liquid and gas phases, respectively; σ – coefficient of surface tension of the liquid; r_1, r_2 – curvature radii of main mutually perpendicular sections of the liquid's surface.

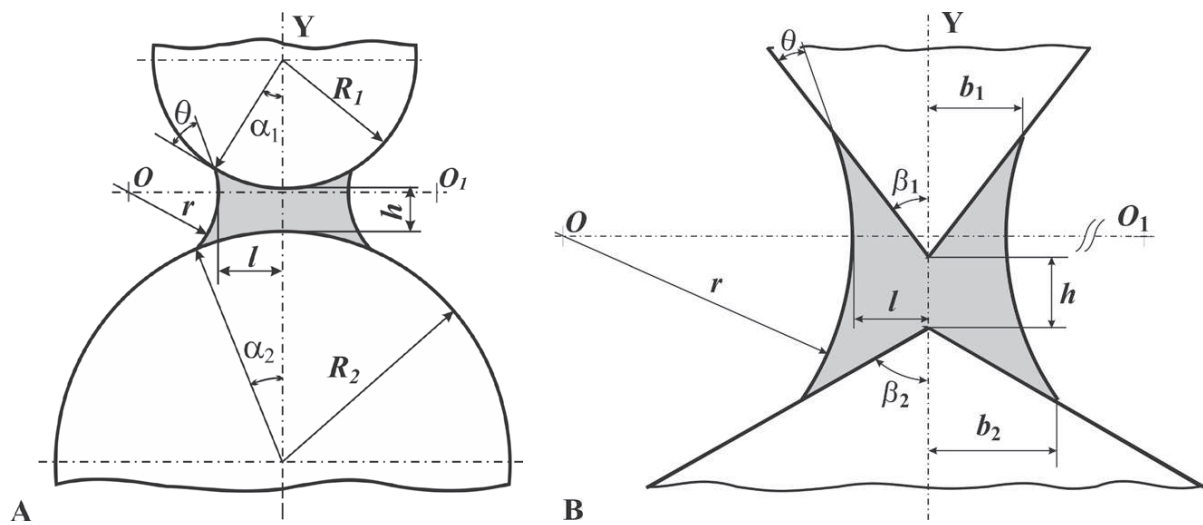


Fig. 1. Computational cell to determine capillary pressure value in liquid interlayer during interaction of sphere-shaped (A) and cone-shaped (B) particles

Microscopic investigations in the shape of the liquid meniscus conducted earlier by us [6], as well as the works by other authors [4, 5, 9, 10] enable approximating the liquid meniscus as a circular arc. With this in mind, the curvature radii of interlayer (r, l) between two interacting spherical particles equal:

$$r = \frac{D_1(1 - \cos \alpha_1) + D_2(1 - \cos \alpha_2) + 2h}{2[\cos(\alpha_1 + \theta) + \cos(\alpha_2 + \theta)]} \quad (2)$$

$$l = \frac{1}{2}D_1 \sin \alpha_1 - r[1 - \sin(\alpha_1 + \theta)] = \frac{1}{2}D_2 \sin \alpha_2 - r[1 - \sin(\alpha_2 + \theta)] \quad (3)$$

here, D_1, D_2 – particle dimensions; α_1, α_2 – angles generated by straight lines: (a) particle center–wetting perimeter, (b) Y axis, and characteriz-

ing the liquid interlayer for both particles, respectively; h – gap between particles; θ – wetting angle.

The liquid's volume is determined as the volume of a figure generated by a circular arc with radius r turning around the vertical axis Y minus volumes of the particles V_c submerged into the liquid:

$$V_l = \pi \int_{y_1}^{y_2} [F(y)]^2 dy - V_c, \quad (4)$$

here, $F(y)$ is the equation of the curve generating surface of revolution around the Y axis; y_1, y_2 – integration limits determined as ordinates of the circumference points wetted by the liquid.

With substituted $F(y)$, V_c , and y_1, y_2 , the eq. (4) can be re-written as:

$$V_l = \pi \left\{ \int_{r \cos(\alpha_1 + \theta)}^{-r \cos(\alpha_2 + \theta)} [r + l - \sqrt{r^2 - y^2}]^2 dy - \frac{D_1^3}{6} \sin^4 \frac{\alpha_1}{2} \left(3 - 2 \sin^2 \frac{\alpha_1}{2} \right) - \frac{D_2^3}{6} \sin^4 \frac{\alpha_2}{2} \left(3 - 2 \sin^2 \frac{\alpha_2}{2} \right) \right\} \quad (5)$$

After replacement of curvature radii (r, l) with expressions (2, 3) in eq. (5), after integration and transformation of eq. (5), the finite expression to calculate the volume of the liquid in interlayer can be obtained. Numerical calculation of eq. (1-5) gives the capillary pressure values in the liquid interlayer.

Results and discussion

Fig. 2 mirrors capillary pressure and the constituent F_p of capillary forces as dependent on the relative volume w during interaction of identical spherical particles and at variable parameters of these particles (here, w is the ratio of the liquid's volume to the interacting particles' volume).

As is apparent from Fig. 2, particle dimensions and wetting angle are basic parameters influencing the capillary pressure value during interaction of identical spherical particles. In accord with the obtained equations, the maximal rarefaction value is attained at zero wetting angle and lesser dimensions of particles. The distance between particles is a less significant factor; at this point, a larger rarefaction is characteristic of a lesser value of h (e.g., the curve 5 as compared with the curve 4, or the converging curves 5, 4 as compared with the curve 3). Variations of the h value, especially at:

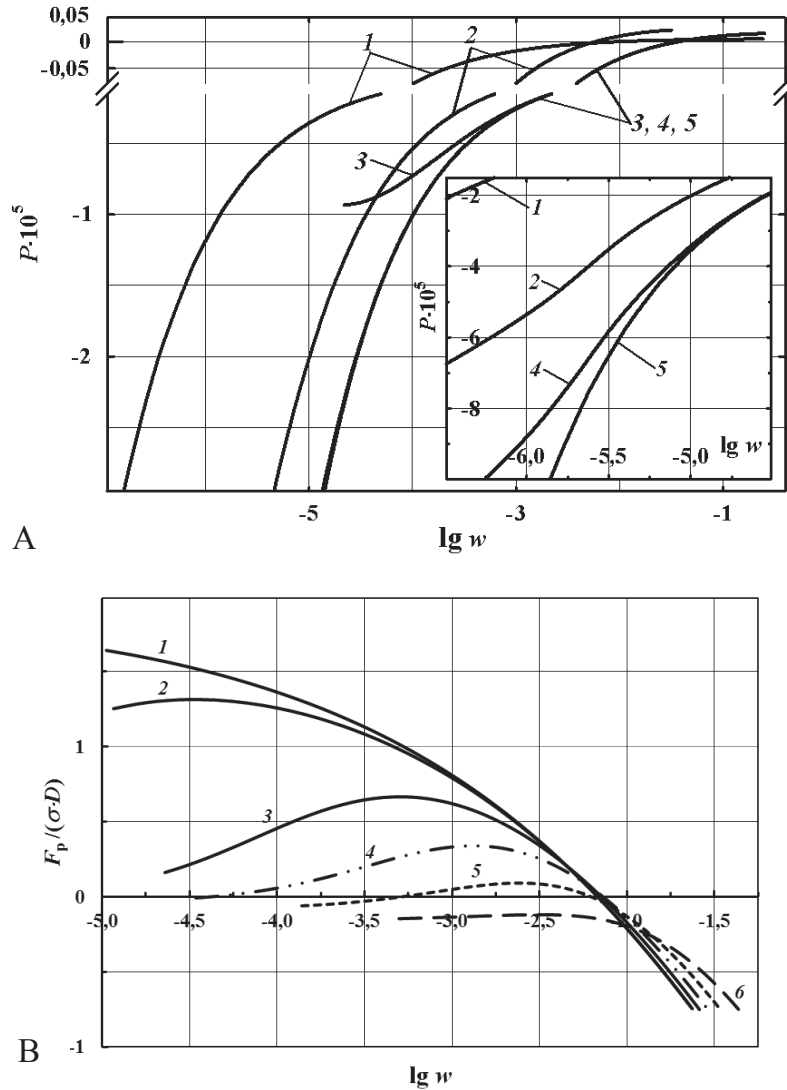


Fig. 2. Capillary pressure in the liquid interlayer (A) and capillary force constituent (B) as dependent on relative volume of interlayer during interaction of spherical particles and at variable (A) D (μm):
 $1 - 100$; $2-5 - 10$; h , μm : $1, 2, 4 - 0.01$; $3 - 0.1$; $5 - 0.2$ nm; θ (degrees): $1, 3-5 - 0$; $2 - 50$; (B) h/D : $1 - 4 \cdot 10^{-4}$; $2 - 0.001$; $3 - 0.01$; $4 - 0.025$; $5 - 0.05$; $6 - 0.1$. $D = 5 \mu\text{m}$, $\theta = 50^\circ \cdot \sigma = 72.8 \text{ mN/m}$

(a) $h < 0.1\%$ of D (curves 4 and 5), and (b) up to 1% (curve 3) are observable only with small portions of the liquid. It appears from this that significant proportions of water contribute to apparition of a repulsive force between the particles. As is apparent also (Fig. 2.B), the increased distance between the particles results in the maximal value of the capillary force constituent F_p shifted to large volumes of the liquid interlayer.

In accord with eq. (1), the capillary pressure value is dependent on the ratio between curvature radii of liquid interlayer. In turn, the ratio between r and l values is determined by values of (a) wetting angle,

and (b) portions of the liquid between the particles. As confirmed by the performed calculations, this ratio follows from the dependence of the interlayer volume on the θ angle (Fig. 3). This dependence is obtained under conditions of zero value of the Laplace pressure within the liquid interlayer between identical spherical particles. Also, this dependence is confirmed by the curves presented in Fig. 2.

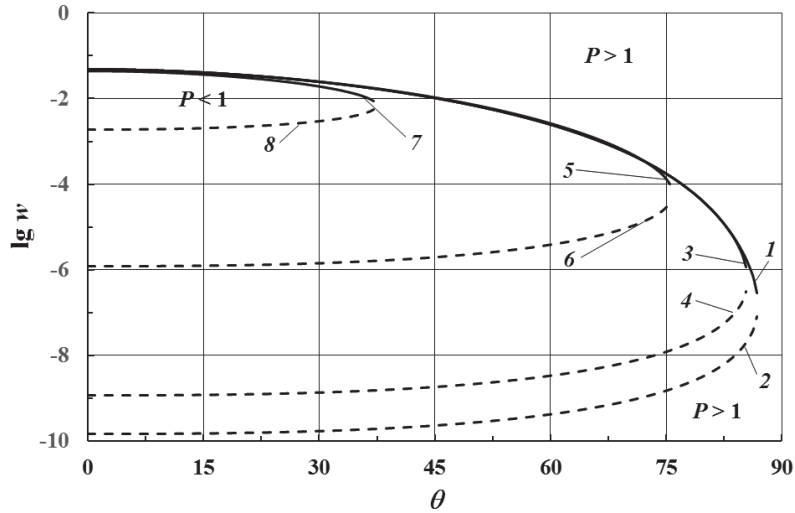


Fig. 3. Relative volume of the liquid in interlayer as dependent on wetting angle at zero capillary pressure and variable gap (h) between particles ($h = \%D$): 1, 2 – 5 nm (0.05); 3, 4 – 10 nm (0.1); 5, 6 – 0.1 μm (1); 7, 8 – 1 μm (10). $D = 10 \mu\text{m}$, $\sigma = 72.8 \text{ mN/m}$

The $P = 0$ condition is realized for two volumes of the liquid interlayer: (a) the minimal (curves 2, 4, 6, 8 in Fig. 3), and (b) the maximal (curves 1, 3, 5, 7 *ibid.*). As is apparent, these curves shape an area featured by capillary rarefaction ($P < 1$). In Fig. 3, $P = 1$ corresponds to 1 atm, or 101325 Pa. Either separation of particles, or the gap between them increased from 0.05% up to 10% of D leads to an appreciable diminution of this area. The poor wetting of the particle surfaces also results in diminution of this area. For values $P > 1$, capillary pressure in the liquid interlayer contributes to repulsion of particles.

In accord with eq. (1-3), the $P = 0$ condition – with identically sized particles – is not dependent on particle dimensions, but is determined by: (a) the portion of liquid in interlayer, (b) wetting angle, and (c) distance between the particle surfaces. Consequently, this condition is appreciably dependent on the properties of particle material, of the liquid, and of the environment.

Fig. 4 reflects how the volume of the liquid requisite to attain the zero value of capillary pressure in the liquid interlayer is dependent on the relative distance between identical spherical particles at the variable wetting angle.

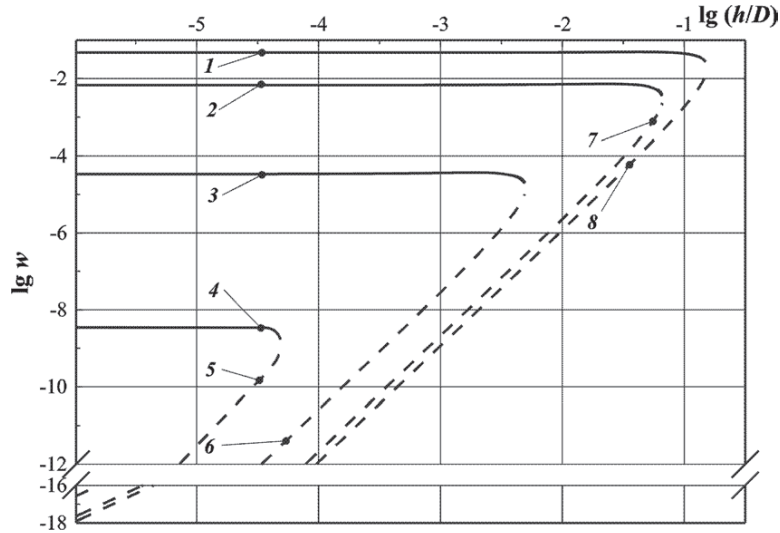


Fig. 4. Relative volume of the liquid interlayer as dependent on relative distance between particles under conditions of $P=0$ and variable wetting angle θ (degrees):
 1, 8 – 0; 2, 7 – 50; 3, 6 – 80; 4, 5 – 89. $D = 5 \mu\text{m}$, $\sigma = 72.8 \text{ mN/m}$

As is apparent, the $P = 0$ condition is attained at two volumes of the liquid: the minimal ($P_{v\min} = 0$, curves 5-8) and the maximal ($P_{v\max} = 0$, curves 1-4). At this point, the area restricted by the mentioned curves containing limit values of the liquid's volumes is featured by rarefaction and, respectively, by maximal value of the capillary force. The poor wetting of the particle surfaces results in diminution of this area and in a narrower diapason of the liquid's volume contributing to apparition of cohesive forces between the particles. For proportions of liquid, both exceeding the maximal (V_{\max}) and lesser the minimal (V_{\min}) values, capillary pressure contributes to repulsion of the particles. At this point, the capillary pressure values in areas over the curves 1-4 ($P_{v\max} = 0$) and below the curves 5-8 ($P_{v\min} = 0$) are determined by both different constituents of the force caused by surface tension of the liquid along its perimeter, and capillary pressure in the liquid interlayer, respectively.

It appeared from the calculation results that basic parameters influencing the capillary pressure value in the liquid interlayer between contacting spherically-shaped particles are the distance between these particles and the wetting angle. At this point, more marked rarefaction is attained under the following conditions: zero wetting angle, lesser distance between the particles, and small volumes of the liquid interlayer. With growing interlayer, the mentioned parameters equalize their influence; also, capillary pressure and the respective capillary force constituent disappear.

Analogically, computational investigations in capillary pressure were performed for the interacting cone-shaped particles. Fig. 5 and 6

demonstrate how capillary pressure is dependent on the relative volume of liquid interlayer w_c during contact of identical cone-shaped particles. (Here, w_c stands for the ratio of the liquid's volume to the volume of the particles submerged into the liquid, at maximal wetting perimeter).

As is apparent from Fig. 5, the cone shape appreciably influences the capillary pressure value. So, with growing angle at the cone vertex, significant capillary rarefaction ($P < 0$) contributing to cohesion of particles is observed. As is noticeable (Fig. 5.A and 5.B), with larger gap

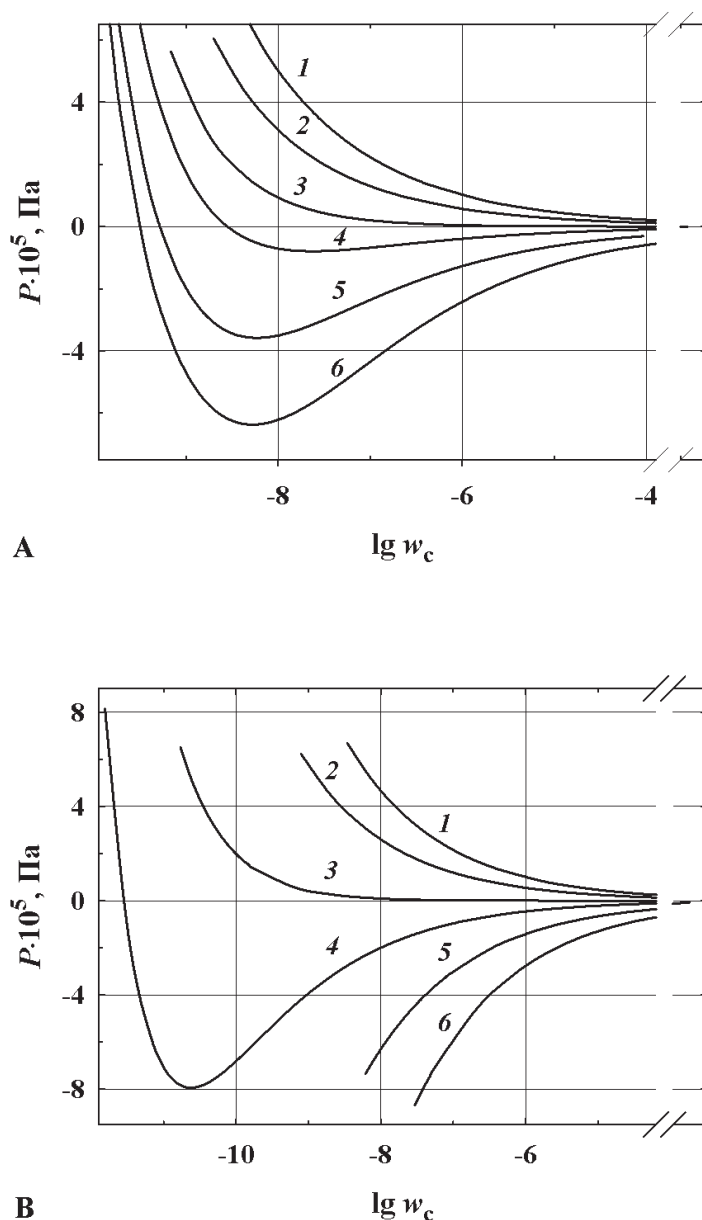


Fig. 5. Capillary pressure in the liquid interlayer as dependent on relative volume of the liquid during interaction of the cone-shaped particles and at variable angle β (degrees) of the cone vertex: 1 – 40; 2 – 80; 3 – 120; 4 – 140; 5 – 160; 6 – 170. $\theta = 0^\circ$, h (mm): A – 0.01, B – 0.001. $\sigma = 2.8$ mN/m

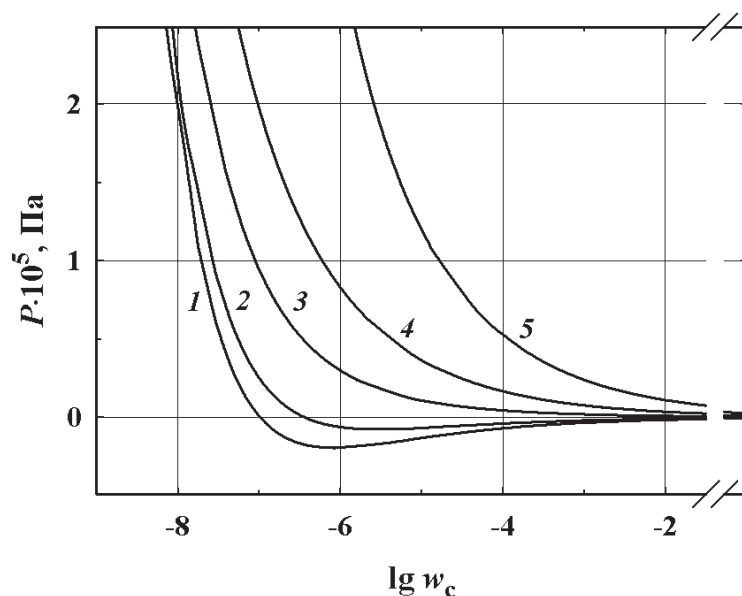


Fig. 6. Capillary pressure in the liquid interlayer as dependent on relative volume of the liquid during interaction of the cone-shaped particles and at variable wetting angle θ (degrees): 1 – 0; 2 – 20; 3 – 40; 4 – 60; 5 – 80. $\beta = 130^\circ$, $h = 0.01 \mu\text{m}$, $\sigma = 72.8 \text{ mN/m}$

between particles, capillary pressure to a larger extent contributes to repulsion of particles ($P > 1$), rather than to their cohesion. The decreased h value leads to the capillary pressure value markedly shifted towards the rarefaction diapason and, respectively, to more intensive coalescence of particles.

As evidenced by the curves presented in Figure 6, the improved wetting contributes to growth of capillary rarefaction and to cohesion of the particles, as is the case with the spherically-shaped particles (Fig. 2-4).

Conclusion

Thus, the offered computational model enables exploring how basic parameters of dispersive particles and of the liquid interlayer influence the capillary pressure value in the interlayer between dispersive component particles. The offered model can be used to numerically investigate how capillary pressure and dispersive component parameters influence the values of cohesive capillary forces.

Acknowledgements

This work was financially supported by the Russian Foundation for Basic Research (Grant Nr. 14-03-00957_a).

References

1. Zimon A.D., Andrianov E.I. *Autoadhesion of Bulk Materials*. Metallurgy Publishers. Moscow. 1978. 287 p.
2. Pietsch W., Haftkraft R. H. *Kapillardruck, Flüssigkeitsvolumen und Grenzwinkel einer Flüssigkeitsbrücke zwischen zwei Kugeln*. Chemie Ingenieur Technik. 1967. H.15. S.885-936.
3. Lykov A.V. *Transfer Phenomena in Capillary-porous Bodies*. The State Publishing House of Technico-theoretical Literature. Moscow. 1954. 296 p.
4. Petzow G., Huppman J.W. *Flüssigphasensintern. Verdichtung und Gefügeausbildung*. Z. Metallkde. 1976. Bd.67. H.9. S.579-590.
5. Eremenko V.N., Naidich Yu.V., Lavrinenko I.A. *Caking in the Presence of Liquid Metal Phase*. Naukova Dumka Publishers. Kiev. 1968. 123 p.
6. Tselishchev Yu. G., Val'tsifer V. A. *Influence of the Type of Contact between Particles Joined by a Liquid Bridge on the Capillary Cohesive Forces*. Colloid Journal. 2003. V.65. N.3. P.385-389.
7. Tselishchev Yu. G. *Evaluation of the Capillary Pressure in a Liquid Interlayer between the Particles of a Dispersed Phase*. Colloid Journal. 2007. V.69. N.4. P.537-539.
8. Tselishev Yu.G., Valtsifer V.A. *Calculation of capillary forces in powder-like materials*. Russian Journal of Chemical Physics and Mesoscopy. 1999. V.1. N.1. P.14-21.
9. Fairbrother R.J., Simons S.J.R. *The rupture energy of liquid bridges between spheres; the effect of contact angle and separation distance on liquid bridge geometries*. World Congress on Particle Technology 3. [in UK] Brighton. 1998. 110 p.
10. Kasai E., Ramos M.V., Kano J., Saito F., Waseda Y. *Simulation model for the agglomeration process of granules during iron ore sintering by discrete element method*. World Congress on Particle Technology 3. [in UK] Brighton. 1998. 89 p.