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Surface tension determination in glyoxal-silica dispersed system

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Abstract. Preliminary studies have shown the possibility of using glyoxal-silica dispersed system for soil stabilization in groundwork base and utility systems. However, the problem associated with optimizing the composition can be solved by measuring energy characteristics. Since the interaction in glyoxal-silica system takes place at the interphase the surface tension was chosen as an energy parameter. The aim of research was to compare calculated by Zisman's and Owens-Wendt-Rabel-Kaelble (OWRK) methods surface tension values of glyoxal-silica composition. Polymineral sand was used as the silica model. The surface tension value calculated by Zisman's method is $\sigma_e=27.47$ mN/m. According to the OWRK method, the polar component of surface tension for test material is $\sigma_s^P=12.61$ mN/m, dispersion component is $\sigma_s^D=13.50$ mN/m, and the total value of surface tension is $\sigma_s=26.11$ mN/m. Comparison of surface tension values calculated by different methods has shown good repeatability. However, the OWRK method provides a more detailed overview of interaction in the glyoxal-silica system and makes it possible to estimate the contribution of polar and dispersion components to the total value of a surface energy characteristic. Therefore, the OWRK method can be recommended for characterizing the structure formation process when optimizing the organomineral compound composition.

1. Introduction

Interfacial processes in coarse, micro and nanodispersed systems are characterized by surface energy changing (E_s). Surface tension (σ) – a free surface energy measure (numerically equal to work of the surface unit formation) – is a criterion that allows E_s value quantification in the structure formation process of the substance. This statement is true for both liquids and solid bodies. However, σ quantitative measurement for solid body surfaces is a difficult experimental task.

There are different methods of surface tension quantitative measurement for solid body surfaces such as "zero creep", crystal cleaving, powder dissolution, "scratch healing", "neutral drop" [1]. But the most effective one is the "neutral drop" method (G.A. Zisman's [2-4] and OWRK (Owens-Wendt-Rabel-Kaelble methods) [5-8]) – a non-destructive method based on measuring the contact angle of solid surface wetting with service fluids.



Zisman's method based on modified Antonov's equation determines the critical value of surface tension (σ_c) for solid surfaces through plotting functional dependency (1) and extrapolating it to $\cos \theta = 1$.

$$\cos \theta = f(\sigma_L) \quad (1)$$

where σ_L is surface tension of service fluids determined experimentally, N/m;

θ is the contact angle of wetting of test material surface.

However, this method has applicability limitations related to the selection process of solution and test structure types. The contact angle of wetting should be measured using low-polar or non-polar liquids. The homologous series of liquids is the most appropriate way. With the liquid surface tension more than 35 mN/m this method generates a high error as it is necessary to take into account the contribution of polar forces to the adhesive interaction value. G.A. Zisman's method is applicable only when the adhesive interaction between contacting phases is determined by dispersion (Van der Waals) forces only, and surfaces have no chemically active centers.

In comparison with the first one, the OWRK method is more universal and based on the fact that free surface energy is a sum of polar and dispersion components (equation 2) and calculated by linear regression (equation 3) plotted based on the results of measuring of the contact angle of solid surface wetting with service fluids with known polar and dispersion elements. Ethylene glycol, ethanol, glycerin and decane with different molecular chemical structure are commonly used as service fluids.

$$\sigma_s = \sigma_s^P + \sigma_s^D \quad (2)$$

where σ_s is surface tension for the test material, N/m;

σ_s^P , σ_s^D are polar and dispersion components of surface tension for the test material, N/m.

$$\frac{\sigma_L(\cos \theta + 1)}{2\sqrt{\sigma_L^D}} = \frac{\sqrt{\sigma_s^P} \sqrt{\sigma_L^P}}{\sqrt{\sigma_L^D}} + \sqrt{\sigma_s^D} \quad (3)$$

where σ_L^P , σ_L^D are polar and dispersion components of surface tension for service fluids, N/m. The values of these elements are shown in the paper [9].

The polar component of the surface energy is formed by means of electrostatic forces originated from polar groups of molecular structures that lost some atoms, and by means of chemical bonds forming electrically neutral dipoles. The dispersion component is defined by Van der Waals interaction formed by the molecular internal energy in the surface layer of material.

The study presented in this paper focuses on comparing calculated by Zisman's and OWRK methods surface tension values of glyoxal-silica composition which is used for soil injection stabilization in groundwork base and utility systems [10, 11].

2. Method

A sedimentary rock – polymineral river sand from the Krasnoflotsky-Zapad deposit (Arkhangelsk region) containing 91% silicon dioxide was used as the silica model.

Sand was preliminarily washed free from clay intrusions and dried to its constant mass at $105 \pm 5^\circ\text{C}$. Grain-size composition is determined by Focused Beam Reflectance Measurement technology using the Lasentec D600E laser particle size analyzer.

The original 40% aqueous solution of glyoxal was diluted to obtain the 20% solution. The 20% aqueous solution of glyoxal was mixed with a mineral component for producing test samples. To obtain the homogeneity of composition the quantity of organic additive has to be 2.3% of sand weight. The resulting mixture having been compacted by the PLG-20 press with a working force of 40 kN, samples were kept in natural conditions at $25 \pm 2^\circ\text{C}$ until the glyoxal polymerization completion which was controlled by measuring the weight of test material. The criterion of polymerization completion was to achieve constant weight.

The DataPhysics OCA 35 automatic unit was used to determine the contact angle of wetting at $20 \pm 2^\circ\text{C}$. Service fluids were aqueous solutions of industrial ethyl alcohol with a water volume ratio from 4 to 50% for Zisman's method and ethylene glycol, ethanol (96%), glycerin and decane for the OWRK method. The contact angle of wetting was determined at during the period of the first contact between the liquid and test material surface which varied within 1.0 ± 0.5 s. This is due to the fact that evaporation, diffusion, capillary phenomena and other processes related to surface irregularity are negligible in this period. The set of experiments consisting of three mandatory parallel measurements with a maximum allowable difference in contact angle of wetting values of $2-3^\circ$ having been completed, functional dependencies (1) and (3) were plotted and the surface tension of test material was calculated.

3. Results

Table 1 contains grain-size composition of sand.

Table 2 contains the surface tension of service fluids and average contact angles of wetting for the glyoxal/silica (sand) system at 2.3% glyoxal content determined by Zisman's method.

Table 3 contains the surface tension of service fluids and average contact angles of wetting for the glyoxal/silica (sand) system at 2.3% glyoxal content determined by OWRK method.

Figure 1 shows functional dependence (1) necessary to determine the critical value of surface tension for the test material at 2.3% glyoxal content.

Figure 2 shows functional dependence (3) necessary to determine the polar and dispersion components and the total surface tension for the test material at 2.3% glyoxal content.

Table 1. Sand particle size distribution obtained by the Lasentec D600E analyzer.

Particle size, μm						
Content, %						
300...500	150...300	50...150	10...50	5...10	1...5	0.5...1
0.27	4.23	28.50	39.69	9.79	17.53	0.00

Table 2. The surface tension of service fluids and average contact angles of wetting for the glyoxal/silica system at 2.3% glyoxal content determined by Zisman's method.

Aqueous solutions of ethanol, % vol.	$\sigma_L \cdot 10^3$, N/m	θ , $^\circ$	$\cos\theta$
96	28.3	8.89	0.988
90	28.7	17.25	0.955
80	29.3	15.42	0.964
70	29.5	21.41	0.931
60	30.6	22.63	0.923
50	30.7	23.79	0.915

Table 3. The surface tension of service fluids and average contact angles of wetting for the glyoxal/silica system at 2.3% glyoxal content determined by OWRK method.

Service fluids	$\sigma_L \cdot 10^3$, N/m	$\sigma_L^P \cdot 10^3$, N/m	$\sigma_L^D \cdot 10^3$, N/m	θ , $^\circ$	$\cos\theta$
glycerin	63.4	26.4	37.0	75.94	0.243
ethanol	21.4	2.6	18.8	8.73	0.988
ethylene glycol	48.8	16.0	32.8	59.31	0.510
decane	23.8	0	23.8	59.60	0.506

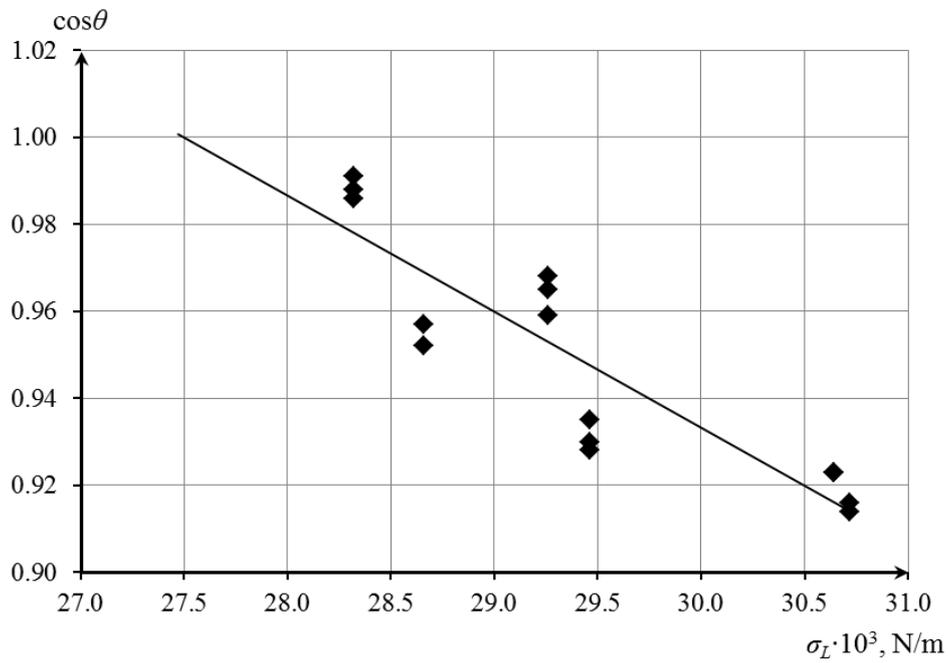


Figure 1. The functional dependence $\cos \theta = f(\sigma_L)$ for the glyoxal/silica system at 2.3% glyoxal content (Zisman's method).

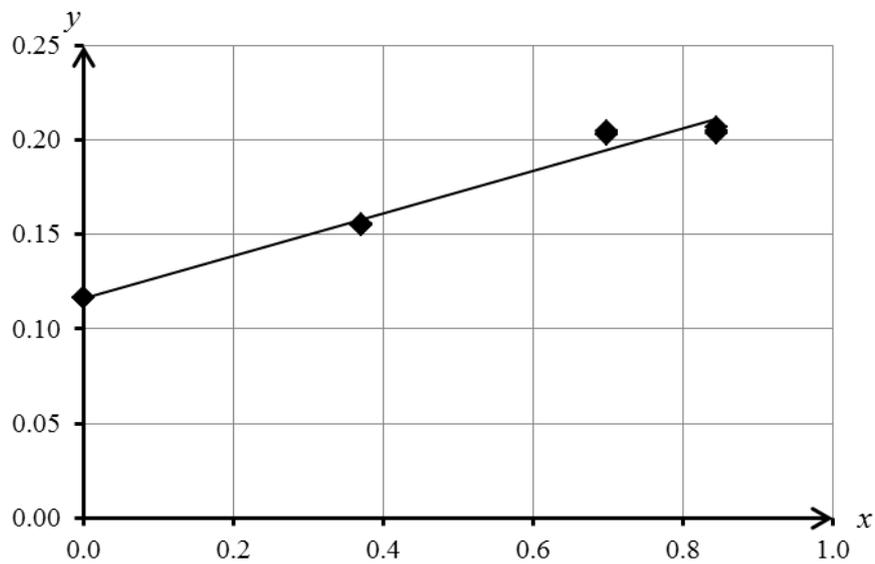


Figure 2. The functional dependence $y = f(x)$ for the glyoxal/silica system at 2.3% glyoxal content (OWRK method).

$$\text{where } y = \frac{\sigma_L(\cos \theta + 1)}{2\sqrt{\sigma_L^D}}; \quad x = \sqrt{\frac{\sigma_L^P}{\sigma_L^D}}$$

4. Discussion

Grain-size analysis of river polymineral sand from Krasnoflotsky-Zapad deposit using the Lasentec D600E particle size analyzer demonstrated that the average size of its grains is 30 μm . Therefore, the investigated silica model is a coarsely dispersed system (particle size exceeds 10^{-5} m). It is found that glyoxal addition does not change the original dispersed structure of sand, but forms a periodic colloidal system by synthesizing the sealing layer of organic polymer that strengthens dispersion interaction between soil particles.

The obtained functional dependencies (Figure 1, 2) are described by linear equations (4, 5) with high values of approximation certainty factor (R), which may indicate a potential for Zisman's and OWRK methods to measure surface tension in the glyoxal-silica dispersed system:

$$\cos \theta = -0.0267\sigma_L + 1.7335, R^2 = 0.8 \quad (4)$$

$$\frac{\sigma_L(\cos \theta + 1)}{2\sqrt{\sigma_L^D}} = 0.1123\sqrt{\frac{\sigma_L^P}{\sigma_L^D}} + 0.1162, R^2 = 0.9 \quad (5)$$

The critical value of surface tension for glyoxal-silica dispersed system calculated by Zisman's method is $\sigma_c = 27.47 \pm 1.15$ mN/m. According to the OWRK method, the polar component of surface tension for test material is $\sigma_S^P = 12.61 \pm 2.02$ mN/m, dispersion component is $\sigma_S^D = 13.50 \pm 0.07$ mN/m, and the total value of surface tension is $\sigma_S = 26.11 \pm 2.67$ mN/m.

In our opinion, comparison of surface tension values calculated by different methods has shown good repeatability despite Zisman's method limitations. Relative value difference does not exceed 5%.

Based on current theories, G.A. Zisman's method allows to determine Van der Waals interaction in the dispersed system only. It should be noted, however, that total surface tension (critical surface tension) experimentally calculated by this method includes both dispersion and polar components. But in comparison with the above, the OWRK method provides a more detailed overview of interaction in the glyoxal-silica system and makes it possible to estimate the contribution of polar (chemical) and dispersion (physical) components to the total value of a surface energy characteristic.

Therefore, the OWRK method can be recommended for characterizing the structure formation process when optimizing the organomineral compound composition.

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