Evaluation of dispersion interaction in glyoxal/silica organomineral system

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Abstract. It is proposed to use an organomineral additive based on glyoxal and saponite-containing waste in order to stabilize the road base. Preliminary studies have shown that soil modification with the organomineral additive with a quantitative ratio of components of glyoxal – 0.52 % and saponite-containing material – 17 % of sand mass increases its specific cohesion by 50 times. However, it is necessary to study the mechanism of interaction between the additive components and soil to optimize the composition and technology of additive introduction into the soil. This paper examines dispersion interaction in glyoxal/silica organomineral system. The Hamaker constant was chosen as a main parameter to evaluate dispersion interaction. This constant is determined based on the principle of measuring the contact angle of wetting with service fluids and calculating the dispersion component of material surface tension. The Hamaker constant in the glyoxal/silica organomineral system was $0.25 \times 10^{-20} – 2.8 \times 10^{-20}$ J at different glyoxal contents, while its maximum value was observed at 0.52 % glyoxal content. The results compared with literature and previously obtained data demonstrated good reproducibility.

1. Introduction

Preliminary studies provided in the paper [1] proved the possibility to use the composite based on glyoxal and saponite-containing waste as an organomineral additive for sandy soil stabilization.

Glyoxal is a dialdehyde of oxalic acid. It is widely used in the industry not only due to the reactivity of its molecules, but also because of the high surface activity of this substance. The additive ensures uniform glyoxal polymerization throughout the soil with a sorbent included in its composition – a saponite-containing waste extracted from tailing dump slurry of industrial ore beneficiation at the diamond deposit named after M.V. Lomonosov.

The organomineral additive components introduced in the soil test samples with a quantitative ratio of glyoxal – 0.52 %, saponite-containing material – 17 % of sand mass increase specific cohesion of the composite by 50 times. The main interaction type of reaction system components is dispersion interaction on the phase boundary in the glyoxal/silica (sand) organomineral system. Currently, however, there is no quantitative estimation of this interaction. Meanwhile, solution of this problem will optimize the composition and technology of additive introduction into the soil.

The objective of studies presented in this paper is to estimate dispersion interaction in the glyoxal/silica (sand) organomineral system in terms of quantity.
The silica model used was a sedimentary rock – polymictic river sand from the Krasnoflotsky-Zapad deposit with 91% of silicon dioxide [2].

The main parameter chosen for the study of dispersion interaction in the glyoxal/silica organomineral system was the Hamaker constant ($A$) which takes into account at least two components acting together – direct interparticle interaction between uniform particles and interface interaction on the solid-solution boundary. The papers [3-5] show that the Hamaker constant is functionally related to surface tension of the dispersion system generated on the phase boundary which characterizes the energy state of the unit of surface.

Surface tension between sand and glyoxal-based organic polymer particles was determined using the Owens, Wendt, Rabel and Kaelble (OWRK) method. The OWRK method rests on the fact that the surface free energy is a sum of polar and dispersive values and determined by linear regression built based on the results of measuring contact angles of wetting with service fluids containing known polar and dispersive components.

The polar component of the surface free 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 dispersive component is defined by Van der Waals interaction formed by the molar internal energy in the material surface layer [3, 6, 7].

According to literature data, the Hamaker constant was already used to estimate dispersion interaction. Thus, [8] gives the following $A$ values for polycrystalline quartz – $4.1 \cdot 10^{-20} \text{–} 4.5 \cdot 10^{-20} \text{ J}$.

2. Method
Sand was preliminarily washed free from clay intrusions and dried to its constant mass at 105±5 °C. The initial 40% aqueous solution of glyoxal was diluted to obtain solutions at a concentration of 1, 2, 5, 10, 20%. Test samples were made by mixing 10 ml of differently concentrated glyoxal solution with 20 g of sand. The resulting mixture was compacted using the PLG-20 press with a working force of 40 kN. Samples were 30 mm in diameter and 14 mm in thickness. Then samples were brought to their constant masses at 40 °C. The constant mass value was taken as a criterion determining the end of glyoxal polymerization.

The contact angle of wetting was determined at the OCA 35L unit. Service fluids were liquids with known polar and dispersive components – water, ethanol, ethylene glycol, decane.

The set of experiments consisting in three mandatory measurements with a maximum allowable divergence of 2-3° contact angle values was followed by calculating the dispersive component of surface tension and the Hamaker constant.

The Hamaker constant for the test material was calculated by the equation (1):

$$A = 24\pi l_0^2 \sigma_S^D$$

where $l_0$ – equilibrium distance between two interacting bodies (particles) which is taken as 0.165 nm for solids; $\sigma_S^D$ – dispersive component of surface tension for the test material.

3. Results
Table 1 contains the surface tension of service fluids and average contact angles of wetting for the glyoxal/silica (sand) system at 0.52% glyoxal content.

Figure 1 shows an example of functional dependence to determine the polar and dispersive components for the test material at 0.52% glyoxal content.

Table 2 contains the polar and dispersive components, total surface tension for the glyoxal/silica (sand) system at different glyoxal weight percentages.

Figure 2 shows the dependence of the Hamaker constant for the glyoxal/silica system on glyoxal content.
Table 1. The surface tension of service fluids and average contact angles of wetting for the glyoxal/silica system at 0.52 % glyoxal content.

| Service fluids   | Surface tension, $\sigma_L \cdot 10^3$, N/m | Polar component of surface tension, $\sigma_L^P \cdot 10^3$, N/m | Dispersive component of surface tension, $\sigma_L^D \cdot 10^3$, N/m | Average contact angle of wetting, $\theta^o$ |
|------------------|---------------------------------------------|---------------------------------------------------------------|---------------------------------------------------------------|--------------------------------------------|
| water            | 72.8                                        | 51.0                                                          | 21.8                                                          | 74.5                                       |
| ethanol          | 21.4                                        | 2.6                                                           | 18.8                                                          | 28.5                                       |
| ethylene glycol  | 48.8                                        | 16.0                                                          | 32.8                                                          | 61.4                                       |
| decane           | 23.8                                        | 0                                                             | 23.8                                                          | 18.8                                       |

Figure 1. The functional dependence $y = f(x)$ for the glyoxal/silica system at 0.52 % glyoxal content.

\[ y = 0.1208x + 0.1102 \]

$R^2 = 0.97$

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

Table 2. The polar and dispersive components, total surface tension for the glyoxal/silica system at different glyoxal weight percentages.

| Glyoxal mass content, $N$, % sand total amount | Polar component of surface tension, $\sigma_S^P \cdot 10^3$, N/m | Dispersive component of surface tension, $\sigma_S^D \cdot 10^3$, N/m | Total surface tension, $\sigma_S \cdot 10^3$, N/m |
|-----------------------------------------------|---------------------------------------------------------------|---------------------------------------------------------------|-----------------------------------------------|
| 0.10                                          | 74.15                                                        | 1.20                                                          | 75.35                                         |
| 0.21                                          | 72.54                                                        | 1.35                                                          | 73.89                                         |
| 0.52                                          | 14.59                                                        | 12.14                                                         | 26.73                                         |
| 1.06                                          | 13.04                                                        | 12.90                                                         | 25.94                                         |
| 2.29                                          | 12.61                                                        | 13.50                                                         | 26.11                                         |
4. Discussion

The Hamaker constant in the glyoxal/silica organomineral system at different glyoxal content was $0.25 \times 10^{-20} - 2.8 \times 10^{-20}$ J. The optimal glyoxal content was taken as 0.52% of sand mass since such an additive content ensures maximum dispersion interaction in the system which does not considerably vary as glyoxal content increases. This fact is in good agreement with the previous studies aimed at determination of physical and mechanical properties of the soil with the organomineral additive.

In our opinion, the Hamaker constant values obtained compared with literature data demonstrated a satisfactory reproducibility. The lower content of this constant in the test sample was caused by inhomogeneity of its structure. Sorbent – saponite-containing material – introduced into the additive contributes to the uniform structure and higher soil strength.

The fact of strengthening Van der Waals interaction between mineral particles of ground in the presence of glyoxal, which was evaluated by Hamaker constant, in our opinion, is connected with forming strong polymer film of the organic component, substituting aqueous dispersion medium. This contributes to the creation of compact periodic colloidal structure (PCS) from monodispersed colloidal silica particles. PCS has a quasicrystalline lattice. Therefore, the physical and mechanical properties of the resulting PCS will be determined by the forces of interaction between the surface of the dispersed particles and the properties of the organic polymer interlayer. The experiments showed to create the such interparticle interlayer in studied organomineral system, a sufficient amount of dialdehyde is 0.52%.

At the same time, the test results indicate that the employed approach can be used to optimize the composition of the organomineral additive.

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