

# THE EFFECT OF PARTICLE SIZE DISTRIBUTION ON APPARENT VISCOSITY OF A DISPERSION SYSTEM

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*Theoretical assumptions on the effect of particle size distribution on apparent viscosity of the system were verified experimentally by measuring the viscosity of polymethyl methacrylate microbeads in glycerine-ethanol mixture and that of ground gypsum in water at various proportions of two or three monodispersional fractions, and at various total concentrations. In terms of proportions of the two particle size fractions, viscosity of a bimodal system exhibits a sharp minimum at otherwise equal total concentration. The effect in question is becoming more pronounced at increasing concentrations. The reduction of viscosity is still more marked with systems comprising three particle size fractions. The course of viscosity vs. composition and the viscosity minimum position established experimentally were in good agreement with the values computed. Mixing of more than three particle size fractions was shown to bring about no substantial decrease in viscosity. The findings can be utilized practically for improving the workability of suspensions, above all mortars, plasters, etc.*

## INTRODUCTION

The effect of particle size distribution on rheological properties of suspensions has not so far been dealt with in any great detail. A number of authors studied the problem from various limited aspects only. For instance, Berens and Hoppe [1] found considerable deviations when measuring the viscosity of lime pastes, and ascribed them to varying particle size distributions. According to Ridge [2], particle size distribution has the greatest effect on the workability of gypsum hemihydrate pastes. The effects of particle size distribution and particle shape were dealt with by Šatava and Škvára [3]. According to their measurements on  $\alpha$  and  $\beta$ -hemihydrate and gypsum, particle size distribution has a considerable effect on the course of apparent viscosity in terms of suspension concentration. The viscosity increases with increasing monodispersity of the system at equal concentration, and the "workability" interval is likewise increasing. Bee [4] found that the viscosity of glass beads-glycerine suspensions decreases with increasing polydispersity.

The effect of size distribution of spherical particles on viscosity of dispersion systems is dealt with in a theoretical paper by Farris [5]. This is based on the assumption that in suspension, small—size particles behave to the larger ones as a liquid. It is further presumed that no interactions between particles of different sizes occur (collisions between particles, formation of structures). According to Farris, this assumption is complied with when the ratio of the grain sizes in question is sufficiently large (at least five-fold). These assumptions then allow to calculate viscosity of a suspension composed

of particles having  $n$  sizes. For relative viscosity  $\eta_r$  of a multimodal system one can derive equation

$$\eta_r = \prod_{i=1}^n H(\Phi_i) \quad (1)$$

where  $H(\Phi_i)$  is a densification factor, and total volume concentration  $\Phi_c$  is given by equation

$$1 - \Phi_c = \prod_{i=1}^n (1 - \Phi_i) \quad (2)$$

where  $\Phi_i$  is volume concentration of  $i$ -th particle size. Farris derived the following equation for a multimodal system on the assumption of zero interaction between the particles:

$$\sum_{i=1}^n \left( \frac{\partial \ln H(\Phi_i)}{\partial \Phi_i} - \frac{(1 - \Phi_n) \partial \ln H(\Phi_n)}{(1 - \Phi_i) \partial \Phi_n} \right) d\Phi_i = 0 \quad (3)$$

Solution of equation (3) can be found when each member of the system is equal to zero. This assumption holds when

$$\Phi_1 = \Phi_2 = \Phi_3 = \dots = \Phi_n \quad (4)$$

and

$$H(\Phi_1) = H(\Phi_2) = H(\Phi_3) = \dots = H(\Phi_n) \quad (5)$$

Solution of equation (3) shows that a suspension of particles having  $n$  sizes exhibits a minimum viscosity at a given concentration and at a certain particle size distribution.

The present study was concerned with further extending and verification of the theory of the effect of particle size distribution on the viscosity of a dispersion system.

#### EXPERIMENTAL

The effect of particle size distribution on viscosity of a dispersion system was first studied on a model suspension [8]. This model was composed of spherical particles suspended in a liquid of equal density, so that the suspension was of virtually Newtonian character. The model comprised polymethyl methacrylate beads in a mixture of glycerine and ethanol. The viscosities were measured of suspensions comprising very narrow bead particle size fractions; their mean diameters were 17  $\mu$ , 100  $\mu$ , 120  $\mu$  and 290  $\mu$ , respectively. The beads were always introduced into the liquid gradually, starting with the smallest ones. Viscosity of the suspensions was measured by means of a rotary viscometer with co-axial cylinders (viscometer RN made in German Democratic Republic) at a constant temperature of 23 °C. The error of viscosity measurement did not exceed  $\pm 5\%$ .

Viscosity measurements on the model suspensions showed that they were practically Newtonian up to a concentration of  $\Phi_c = 0.55$ . The effect of flow limit appears at higher concentrations (Fig. 1). The dependence of suspension viscosity on concentration was measured for 100  $\mu$  particles (Fig. 2). Fig. 2 also shows a plot of viscosity concentration according to relations suggested earlier (cf. Appendix 2).

The Effect of Particle Size Distribution

At concentrations  $\Phi_c = 0.3 - 0.6$  the suspension viscosity was measured at various contents of fine ( $17 \mu$ ) and coarse ( $120 \mu$ ) particle size fractions. Theoretical viscosity courses were computed for the same contents of fine

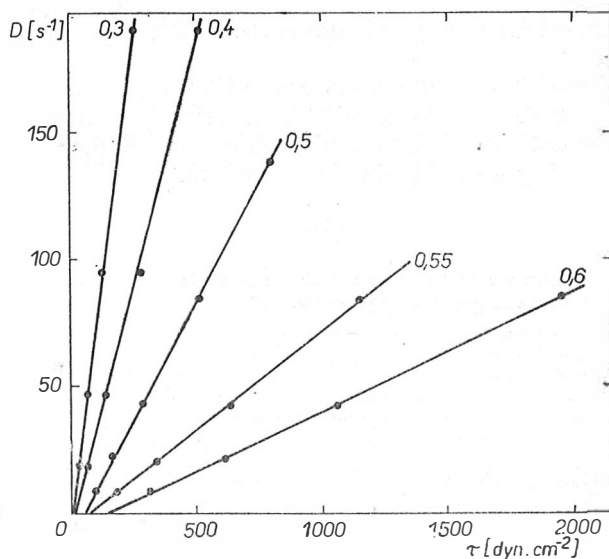


Fig. 1. Velocity gradient and stress in suspensions of PMMA spheres in glycerine-ethanol mixture at various volume concentrations.

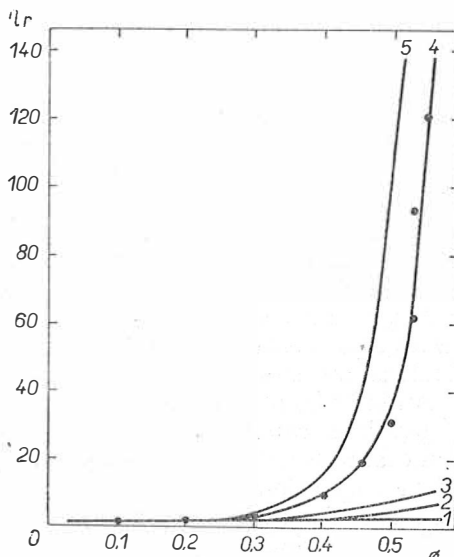


Fig. 2. Relationship between relative viscosity and volume concentration for suspensions of PMMA beads in glycerine-ethanol mixture; • — experimental values, curve according to 1 — Einstein 2 — Brinkman, 3 — Farris, 4 — Lee 1, 5 — Lee 2.

and coarse fractions (cf. Appendix 1; Fig. 3). The viscosities of trimodal systems were measured at concentration  $\Phi_c = 0.5$  (fine fraction 17  $\mu$ , mean fraction 100  $\mu$ , coarse fraction 280  $\mu$ ). With this volume concentration the lowest viscosity was found at a composition of 34 % by vol. of fine, 40 % by vol. of medium and 26 % of coarse fraction. The region of minimum viscosity thus found was in good agreement with the computed viscosity values (Fig. 4).

The lowest viscosities found at concentration  $\Phi_c = 0.5$  for bimodal and trimodal systems were compared with theoretically computed values. The calculation of theoretical minimum viscosities was likewise carried out for suspensions comprising 4 to 6 grain size fractions.

Table I

Relationship between relative viscosities of aqueous suspensions of gypsum and particle size composition of the solid

Particle size fraction		Viscosity [ $P$ ]	
Coarse	Fine	50 % by vol.	58 % by vol.
10%	90%	Suspension is strongly dilatant	Suspension is strongly dilatant
20	80		
30	70		
40	60		
50	50		
60	40	19.1	29.8
70	30	14.2	16.1
75	25	9.2	12.3
80	20	5.8	7.5
90	10	9.4	14.2

The results obtained by measuring the viscosity of the model system were supplemented by measuring the viscosity [17] of ground gypsum in water. Aqueous suspensions of ground gypsum also represent a model system which, however, is more similar to real suspensions. The particles of ground gypsum are not spherical, being corpuscular, and their density is different from that of water. The suspension ceases to settle only after attaining a certain concentration of the solid which forms a certain internal structure. The rheological character of gypsum suspensions is strongly un-Newtonian. The apparent viscosities were also measured with a rotary viscometer. However, viscosity is very difficult to measure at very high concentrations of solids so that qualitative data can only be obtained. The results gained from measurements of gypsum suspensions can be applied to hemihydrate suspensions which are difficult to measure (setting of the suspension).

In the case of gypsum suspensions, apparent viscosity of bimodal systems (fine fraction 5—20  $\mu$ , coarse fraction 100—120  $\mu$ ) and of trimodal ones (fine fraction 5—20  $\mu$ , medium fraction 63—100  $\mu$ , coarse fraction 160—200  $\mu$ ) was measured. With bimodal systems, similarly to the model suspension the viscosity minimum was found at a composition of 20—30 % by vol. of fine fraction, and 80—70 % of coarse fraction (Table I). At this grain size

distribution it is still possible to process suspensions containing 63 % by vol. of solid. The viscosity minimum for trimodal system was established at a composition of about 46 % by vol. of coarse fraction, 32 % by vol. of medium fraction and 22 % by vol. of fine fraction. At this composition it is still possible to process aqueous gypsum suspensions containing 66 % by vol. of solid phase. At other particle size compositions such suspensions cannot even be prepared.

A series of programs in the Algol and Fortran languages has been worked out for computing the theoretical viscosity of optimum suspensions. The computing proper was carried out with computers NCR Elliot 4120 [10], Hewlett—Packard 2116 B [9], and Tesla 200 [11].

### DISCUSSION

The experimentally established relationship between relative viscosity and volume concentration for a monomodal system of polymethyl methacrylate beads in glycerine-ethanol mixture was compared with several similar relationships described in literature. At low volume concentrations of solid phase one can observe a very good agreement of all the suggested equations with experimental data. At higher concentrations there appear substantial deviations. The values measured agree best to the equation suggested by Lee [4] (cf. Appendix 2, equation Lee 2). This equation was taken as basis for reading the viscosities of the individual fractions.

The relationship between relative viscosity and composition of suspension in the case of bimodal suspension of methyl methacrylate beads in glycerin-alcohols mixture, as well as in that of aqueous gypsum suspensions, shows a marked decrease in viscosity at a given concentration. The sharpness of the minimum increases with increasing concentration. Fig. 3 shows that at total concentration  $\Phi_c = 0.55$  a mixture of two fractions at a ratio of 30 %

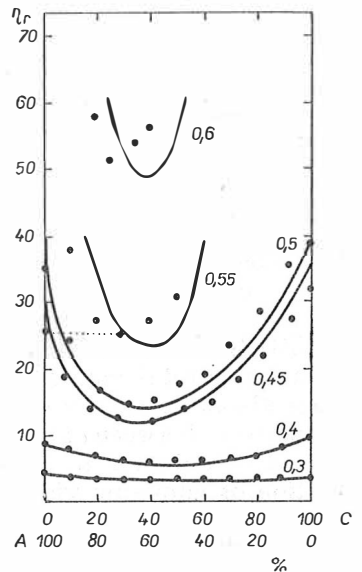


Fig. 3. Plot of relative viscosity vs. particle size composition of the solid in binary system; A — coarse particle size fraction, C — fine particle size fraction, ● — experimental values (PMMA beads in glycerine-ethanol mixture), — theoretically computed values.

by vol. of fine and 70 % by vol. of coarse, has virtually the same viscosity as a suspension of one fraction at lower volume concentration  $\Phi_c = 0.45$ . Mixtures having a high content of solid phase ( $\Phi > 0.6$ ) can be pasted only at certain proportions of particle size fractions only. At higher concentrations the values determined experimentally are also shifted with respect to computed viscosity values. This shift is probably due to a failure to comply with the assumption of zero interaction between particles in suspension, as well as due to the non-Newtonian character of the suspensions. The experimental data obtained for the bimodal system (Fig. 3) show a shift of minimum viscosities towards a lower percent content of fines at higher total concentrations of solids. The relative viscosity minima in a bimodal system roughly

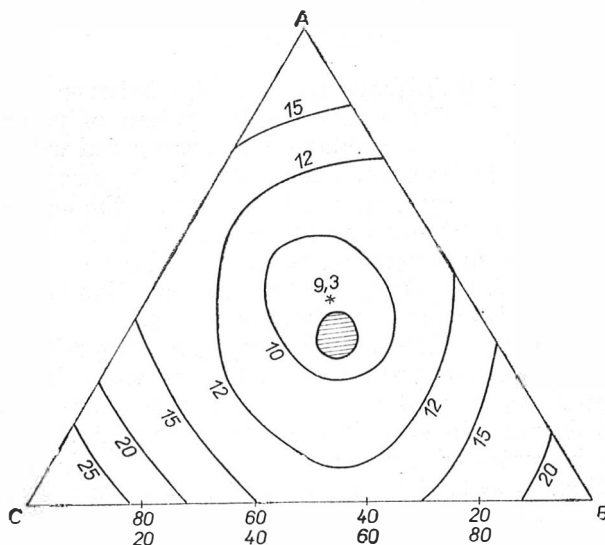


Fig. 4. Relative viscosity vs. particle size composition of the solid in a trimodal system at total volume concentration 0.5; A — coarse particle size fraction, B — middle fraction, C — fine particle size fraction, \* — viscosity minimum, • — region of experimentally established viscosity minimum.

comply with the theory of combining maximum density solids [18]. This theory assumes that the gaps between primary (large) spheres are filled by secondary (small) spheres. This theory does not hold accurately for the system in question as it does not allow to explain the shift of minimum viscosity when total concentration of the suspension is increased.

With trimodal systems a still higher decrease in viscosity is attainable at the same total concentration. Increasing concentrations of the suspensions bring about increasing sharpness of the viscosity minimum, as shown by comparison between Figs. 4 and 5. Increasing concentration causes the viscosity minimum to be shifted towards a lower content of fines. The position of minimum viscosity regions is in good agreement with the values computed. In the case of aqueous gypsum suspensions the authors succeeded in preparing a workable suspension at a solid phase content approaching

the theoretical water coefficient for hemihydrate suspensions, by mixing three grain size fractions at proportions corresponding to the viscosity minimum.

The viscosity of optimized suspensions decreases with increasing number of particle size fractions. The theoretical viscosity values approach a limit value with the increasing number of grain size fractions (Fig. 6). Raising the number of fractions above 3 brings about no further decrease in viscosity of the optimized system. In addition to this, perfect mixing of more than three fractions is not always feasible in actual practice.

The experiments as well as computations have borne out the assumptions of viscosity reduction at suitable granulometric composition. The assumption

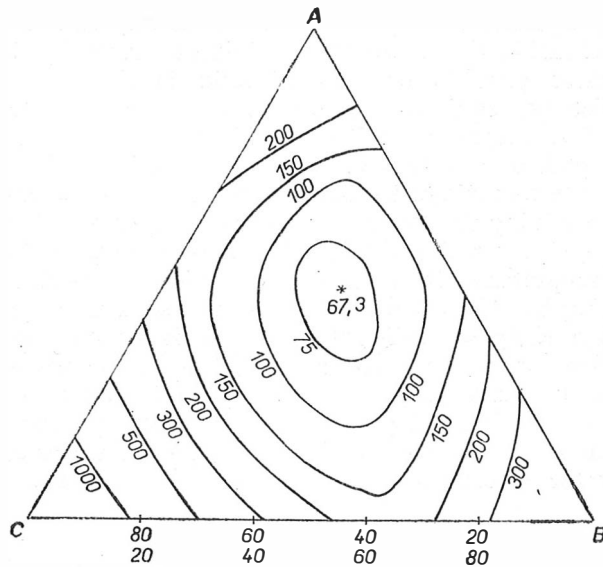


Fig. 5. Relationship between relative viscosity and particle size composition of the solid in a trimodal system at total volume concentration 0.66; A — coarse particle size fraction, B — middle fraction, C — fine particle size fraction, \* — viscosity minimum.

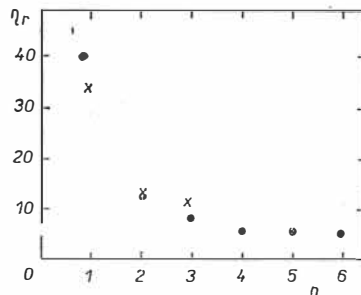


Fig. 6. Minimum relative viscosity in optimized systems vs. the number of solid particle size fractions ( $n$ ); x — experimental viscosity values for suspensions of PMMA beads in glycerine-ethanol mixture. ● — theoretically established values.

of zero interaction between particles of various size is met at low concentration of the solid only. At higher concentrations the computed viscosity values comply with experiments to a qualitative degree only. However, a decrease in viscosity was found to take place at a suitable particle size composition of the solid not only in the case of spherical-shaped particles, but also with non-spherical ones. By selecting a suitable composition of solid it is possible either to improve workability of the suspension at a given concentration of the solid, or to increase the content of the solid in suspension at a certain consistency of the suspension.

#### CONCLUSION

Rheological properties of suspensions are frequently of decisive significance in technological utilizations, where a minimum viscosity is often sought while the highest possible content of solid is simultaneously required. These contradictory requirements are usually met by a compromise, i.e. use is made of a suspension with the highest proportion of solids, still exhibiting acceptable consistency. Workability of a suspension does not depend on the volume ratio of the solid only, but also on a number of further parameters determining the "structure" of the suspension, in particular with respect to shape, size and size distribution of the solid particles. By adjusting suitably these magnitudes it is possible to achieve an optimum workability while maintaining the given volume ratio of the solid in suspension.

Increasing the share of solids in mortars is of considerable practical significance for increasing the strength of hardened suspensions. The principle of suitable granulometry [12] for preparing materials of high strength is applicable for suspensions of gypsum hemihydrate, portland cement, etc. The findings can also be utilized in the conveying of slurries or sludges where a minimum water content and maximum fluidity of the suspension are required.

#### APPENDIX 1

Equations (1) and (2) were derived on the assumption that in a suspension of spherical particles the fines behave to the coarse particles as a liquid does. Viscosity of fine particles in a liquid can be defined as follows:

$$\eta_r = \frac{\eta_j}{\eta_0} = H(\Phi_j)$$

where  $\eta_0$  — is viscosity of the liquid  
 $\eta_r$  — is relative viscosity  
 $H(\Phi_j)$  — is the densification factor.

The suspension is thickened (densified) when coarse particles are added to a suspension of fine particles. The factor of densification due to the coarse particles is defined

$$H(\Phi_h) = \frac{\eta_h}{\eta_j}$$

where  $\eta_h$  is viscosity of suspension composed of coarse particles  
 $\eta_j$  is viscosity of suspension composed of fine particles.

The densification factor is in fact the contribution of relative viscosity of the particle size fraction added to the suspension. Relative viscosity of a mixture of fine and coarse particles in suspension in a liquid can be defined by the expression

$$\eta_r = H(\Phi_h) \cdot H(\Phi_j)$$

Generally, a system composed of particles having  $n$  sizes is defined

$$\eta_r = \prod_{i=1}^n H(\Phi_i)$$

where the particle size increases with increasing index from 1 to  $n$ .

Volume concentration of fine particles in suspensions  $\Phi_j$  is defined

$$\Phi_j = \frac{V_j}{V_0 + V_j}$$

where  $V_j$  is the volume of fine particles

$V_0$  is the volume of the liquid.

Volume concentration of coarse particles in a suspension of coarse and fine particles in a liquid is

$$\Phi_h = \frac{V_h}{V_h + V_j + V_0}$$

Generally, the following equation holds for a system of particles having  $n$  sizes:

$$\Phi_i = \frac{V_i}{\sum_{i=0}^n V_i} \quad \Phi_c = \frac{\sum_{i=1}^n V_i}{\sum_{i=0}^n V_i} \quad (6)$$

where  $\Phi_i$  is the concentration of  $n$ -th size particles

$\Phi_c$  is total concentration.

From equations (6) it follows that

$$(1 - \Phi_c) = \prod_{i=1}^n (1 - \Phi_i) \quad (7)$$

Equation (7) and conditions (4) and (5) allow to compute the composition of a multimodal system which has a minimum viscosity at given total concentration  $\Phi_c$ . In that case, equation (7) is solved for the given  $n$  at the given total concentration.

Equation (7) was solved for  $n = 2$  to 6 and for various  $\Phi_c$  using the method of gradual approximations. The volumes of optimized composition were then computed according to equation (6). The relative suspension viscosity was computed according to relation (1). The relative viscosities of the individual particle size fractions, which are equal to densification factors  $H(\Phi_i)$ , were read off the experimental dependence of relative viscosity on concentration (Fig. 2). Reading of relative viscosities from the dependence of  $\eta_r$  on concentration with a monodispersion system for various sizes of spherical particles is possible because the relation between  $\eta_r$

and concentration is independent of the size of spherical particles [4, 5, 13—16].

The above relationships also allow to compute relative viscosities of suspensions at given particle size compositions. Known volumes  $V_i$  and  $V_0$  at known  $\Phi_c$  permit the respective  $\Phi_i$  to be calculated easily, and by determining relative viscosity for the computed  $\Phi_i$  from relationship between  $\eta_r$  and concentration, to compute the relative viscosity of the suspension. This method was used in computing isocomes for a bimodal and a trimodal system at various total suspension concentrations.

## APPENDIX 2

A number of equations have been suggested for the relation between relative viscosity  $\eta_r$  and volume concentration  $\Phi$ . The following equations were employed in the present study:

$\eta_r = 1 + 2.5 \cdot \Phi$	Einstein [6]
$\eta_r = (1 - \Phi)^{-2.5}$	Brinkman [7]
$\eta_r = (1 - \Phi)^{-3}$	Farris [5]
$\eta_r = 1/(1 - \Phi)^{(2.5 + 1.92 \cdot \Phi + 7.739 \cdot \Phi^2)}$	Lee 1 [4]
$\eta_r = 1/(1 - \Phi)^{(2.5 + 3.25 \cdot \Phi + 12.253 \cdot \Phi^2)}$	Lee 2 [4].

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VLIV ROZDĚLENÍ VELIKOSTI ČÁSTIC NA ZDÁNLIVOU  
VISKOZITU DISPERZNÍHO SYSTÉMU

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Theoretické předpoklady o účinku rozdělení velikostí částic na zdánlivou viskozitu systému byly experimentálně prokázány měřením viskozity suspenzí dentakrylových

кулиček v glycerinalkoholu a mletého sádrovce ve vodě. Měření byla prováděna při různém poměru dvou nebo tří monodisperzních frakcí a při různé celkové koncentraci. Viskozita v bimodálním systému vykazuje v závislosti na poměru obou frakcí ostré minimum při stejné celkové koncentraci. Se vzrůstající koncentrací je tento efekt výraznější. Snížení viskozity je ještě ztelnější u systému složeného ze tří frakcí. Průběh viskozit v závislosti na složení a poloze minima viskozity zjištěné experimentálně byly v dobré shodě s vypočtenými hodnotami. Teoreticky bylo prokázáno, že smíšení většího počtu frakcí než tři nepřináší další podstatné snížení viskozity. Získaných poznatků je možno prakticky využít pro zlepšení zpracovatelnosti suspenzí maltovin i jiných suspenzí.

- Obr. 1. *Závislost rychlostního gradientu a napětí suspenzí dentakrylových kuliček v glycerinalkoholu při různých objemových koncentracích.*
- Obr. 2. *Závislost relativní viskozity na objemové koncentraci pro suspenzi dentakrylových kuliček v glycerinalkoholu; • — experimentální hodnoty, křivka 1 — Einstein, 2 — Brinkman, 3 — Farris, 4 — Lee 1, 5 — Lee 2.*
- Obr. 3. *Závislost relativní viskozity na složení pevné látky v bimodálním systému; A — hrubá frakce, C — jemná frakce, • — experimentální hodnoty (dentakrylové kuličky v glycerinalkoholu, — teoreticky vypočtené hodnoty.*
- Obr. 4. *Závislost relativní viskozity na složení pevné látky v trimodálním systému při celkové objemové koncentraci 0,5; A — hrubá frakce, B — střední frakce, C — jemná frakce, \* — minimum viskozity, • — oblast experimentálně nalezené minimální viskozity.*
- Obr. 5. *Závislost relativní viskozity na složení pevné látky v trimodálním systému při celkové objemové koncentraci 0,66; A — hrubá frakce, B — střední frakce, C — jemná frakce, \* — minimum viskozity.*
- Obr. 6. *Závislost minimální relativní viskozity v optimalizovaných systémech na počtu frakcí pevné látky (n); × — experimentální hodnoty viskozit suspenzí dentakrylových kuliček v glycerinalkoholu, • — teoreticky vypočtené hodnoty.*

## ВЛИЯНИЕ РАСПРЕДЕЛЕНИЯ ЧАСТИЦ ПО РАЗМЕРУ НА „КАЖУЩУЮСЯ“ ВЯЗКОСТЬ ДИСПЕРСНОЙ СИСТЕМЫ

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Теоретические предположения о влиянии распределения частиц по размеру на кажущуюся вязкость системы были доказаны на основании измерения вязкости суспензии дентакриловых шариков в глицериналкоголе и измельченного известняка в воде. Измерения проводили при разном соотношении двух или трех монодисперсных фракций и при разной общей концентрации. В зависимости от соотношения обеих фракций и при одинаковой общей концентрации вязкость в бимодальной системе дает резкий минимум, который в зависимости от нарастающей концентрации становится более выразительным. В случае системы, состоящей из трех фракций окажется понижение вязкости еще более выразительным. Ход вязкости в зависимости от состава и минимума вязкости, установленные экспериментальным путем, находятся в хорошем согласии с рассчитанными величинами. Теоретически было доказано, что смешивание большего количества фракций, чем три, не вызывает дальнейшего существенного понижения вязкости. Полученные результаты можно практически использовать для улучшения приготовления растворов и других суспензий.

Рис. 1. *Зависимость градиента скорости и напряжения суспензий дентакриловых шариков в глицериналкоголе при разных концентрациях по объему.*

Рис. 2. *Зависимость относительной вязкости от концентрации по объему для суспензий дентакриловых шариков в глицериналкоголе; • — экспериментальные величины, кривая 1 — Эйнштейн, 2 — Бринкман, 3 — Фаррис, 4 — Ли 1, 5 — Ли 2.*

- Рис. 3. Зависимость относительной вязкости от состава твердого вещества в бимодальной системе; А — крупная фракция, С — мелкая фракция, ● — экспериментальные величины (дентакриловые шарики в глицериналкоголе), — теоретически рассчитанные величины.
- Рис. 4. Зависимость относительной величины вязкости от состава твердого вещества в тримодальной системе при общей концентрации по объему 0,5; А — крупная фракция, В — средняя фракция, С — мелкая фракция, = — минимум вязкости, ○ — область экспериментальными путем найденной минимальной вязкости.
- Рис. 5. Зависимость относительной вязкости от состава твердого вещества в тримодальной системе при общей концентрации по объему 0,66; А — крупная фракция, В — средняя фракция, С — мелкая фракция, \* — минимум вязкости.
- Рис. 6. Зависимость минимальной относительной вязкости в оптимизированных системах от количества фракций твердого вещества ( $n$ ); ● — экспериментальные величины вязкости суспензий дентакриловых шариков в глицериналкоголе, — теоретически рассчитанные величины.