

## EFFECT OF STRUCTURAL VISCOSITY ON THE PROPAGATION OF ULTRASONIC WAVES IN DILUTE SUSPENSIONS

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The aim of the paper is a rigorous derivation of the formulas determining the phase velocity and the attenuation coefficient of ultrasonic waves propagating in diluted suspensions, and experimental verification of these theoretical predictions. In experiments, the ultrasonic waves were propagating through the suspension composed of peat suspended in water. The satisfactory fitness of the values implied by the theoretically obtained formulas and the respective experimental data allow us to recommend the ultrasonic method as a non-invasive, rapid, accurate and cheap method that might be used for assessments of the solid content in suspensions.

### 1. Introduction

This paper deals with the modeling of the attenuation of ultrasonic waves in diluted suspensions and with the experimental verifications of the theoretical predictions. The problem is considered for the case where the length of the ultrasonic wave is enough large to allow us to neglect the scattering contribution to the attenuation of the ultrasonic wave. In the present considerations, only the loss of the mechanical energy following from viscous flows in the acoustic field of the ultrasonic wave contributes to the attenuation.

The main purpose of the present studies is to test the ultrasonic method as one of the non-invasive methods (see OBRAZ 1983; MALCOLM and POVEY 1997) suitable for determination of the volume fractions of the suspensions under examination, i.e. the ratio of the solid particles volume to the total volume of the medium as a whole. The investigation of the ultrasonic wave propagation in suspensions, in particular, the measurement of wave velocity and attenuation enables us to perform a rapid, relatively cheap and accurate assessment *on line* of the solid volume concentration in the suspension under examination.

The problem considered here is not a new one (see e.g. ACHENBACH and ZHANG 1990; AHUJA 1972; AUZERAIS *et al.* 1988; BERGSTRÖM 1992; CLEMNET *et al.* 1990; HARKER and TEMPLE 1987; LEWANDOWSKI 1992), however, the theoretical approach

to determination of the phase velocity and attenuation coefficient of the ultrasonic waves presented here differs from those proposed in the mentioned papers.

The present studies concern dilute suspensions of solid particles in water (of solid fraction up to about 0.20), for which the assumption of uniform distribution of solid particles throughout the solvent is valid. We shall consider such a medium as a two-phase mixture of liquefaction consistency, which can be modeled by single compressible liquid of structural *viscosity*\*. The motion of the mixture disturbed by the propagating ultrasonic waves may be described by the Navier-Stokes equation and the equation of mass continuity for the mixture as a whole. This motion can be consider for small fluctuations (displacements) of the liquid particles from their equilibrium positions. Also the region, in which the disturbances occur during the ultrasonic measurements, is small since the cross-sections of ultrasonic heads used for measurements were of small dimensions in our studies.

The relative motion of solid particles with respect to viscous solvent, causing the viscous drag of the particles motion, is taken into account in derivation of the structural viscosity. We assume that each particle contributes to the local resistance of motion (drag forces). Calculating the drag force, we derive the formula for structural viscosity, which is an evident (nonlinear) function of volume fraction, different from that presented in BERGSTRÖM (1994), HARKER and TEMPLE (1987), or BANDROWSKI *et al.* (2001).

The experimental investigations were carried out on the suspensions made of peat and water. The ultrasonic measurements were performed in a special tub filled up with this suspension, in which the ultrasonic heads were immersed (measurement *in situ*). The time of propagation of ultrasonic waves was measured with the help of ultrasonic tester type UMT-01 produced by UNIPAN (see KOWALSKI and SIKORSKI (2002)).

The theoretical predictions fit pretty well the experimental data for suspensions of solid volume fraction less than 20%, and the smaller the volume fraction of the solid particles, the better the fitness. For greater volume fractions, another model for the sediment ought to be used.

## 2. Determination of the phase velocity and the attenuation coefficient of an ultrasonic wave in a dilute suspension

The objective of this study is to estimate the volume fraction of the solid phase in a suspension or the density profile of the sediment by acoustic technique. In this technique, either the phase velocity or the attenuation coefficient formulas, implied by the suitable experimentally determined physical coefficients, may be useful in

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\* The structural viscosity is called also the relative viscosity (BERGSTRÖM, 1994), or effective velocity (HARKER and TEMPLE, 1998), or apparent viscosity (BANDROWSKI *et al.* 2001).

determining of the solid content in suspensions. The solid volume fraction is involved in the formulas for phase velocity and attenuation coefficient, so it can be estimated very accurately from the measurements of these quantities in real suspensions, after a suitable calibration of measure setup.

In order to derive the formulas for the phase velocity and the attenuation coefficient, we assume what follows:

- The considerations concern diluted suspensions of volume fraction less than 20%.
- The suspension is considered for a two-phase mixture with continuously distributed solid particles in the liquid solvent, Fig. 1.
- The liquid (solvent) is characterized by the dynamic viscosity  $\mu$ , while the liquefied mixture (suspension) by the structural (effective) viscosity  $\mu_s$ ; the relation between them is given in the next section.
- The ultrasonic waves are regarded to be small fluctuations of the displacements (oscillations) of the medium particles, the oscillations being harmonically dependent on the position vector and time (with a constant frequency).
- The ultrasonic waves are assumed to be plane and linearly polarized in the propagation direction  $0-x$ .
- The motion of the mixture as a whole is described by the Navier-Stokes equation in terms of the so called structural viscosity.
- The relative motion of solid particles with respect to viscous solvent, causing the viscous drag force on the particles, is taken into account in derivation of the structural viscosity.

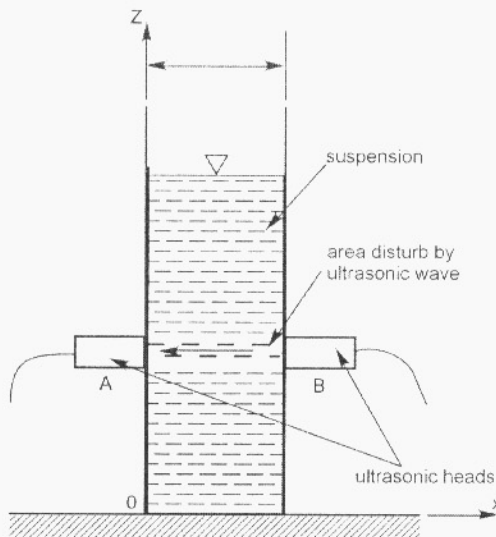


Fig. 1. Suspension in a sedimentation column with attached ultrasonic heads.

• The heat transfer between solid and liquid phases during ultrasonic wave propagation is neglected.

• The solid and liquid as a single media are incompressible, but their mixture is compressible due to variation of the volume fraction.

• The pressure of the mixture is an explicit function of its density.

Let  $\rho^{sr}$  and  $\rho^{lr}$  denote the real mass densities of solid particles and liquid (solvent),  $\phi$  is the volume fraction of the solid particles,  $\rho^s = \rho^{sr}\phi$  and  $\rho^l = \rho^{lr}(1 - \phi)$  are the partial mass densities for solid and liquid, and  $v^s$  and  $v^l$  denote the velocities of solid and liquid particles in the direction of ultrasonic wave propagation coinciding with x-direction. While both modeled media are regarded to be continuously distributed through the space, we are allowed to write the equations of mass continuity for these individual constituents in the form

for solid

$$\frac{\partial \rho^s}{\partial t} + \frac{\partial \rho^s v^s}{\partial x} = 0, \quad (1)$$

for liquid

$$\frac{\partial \rho^l}{\partial t} + \frac{\partial \rho^l v^l}{\partial x} = 0. \quad (2)$$

Adding these two equations, we get the mass continuity equation for the mixture as a whole

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho v}{\partial x} = 0, \quad (3)$$

where  $\rho = \rho^s + \rho^l$  is the mass density of the mixture as a whole, and  $v = (\rho^s v^s + \rho^l v^l)/\rho$  is the average velocity of motion of the mixture as a whole.

The one-dimensional Navier-Stokes equation for the mixture as a whole may be written in the form

$$\rho \left( \frac{\partial v}{\partial t} + \frac{\partial v}{\partial x} v \right) = - \frac{\partial p}{\partial x} + \frac{4}{3} \mu_s \frac{\partial^2 v}{\partial x^2}, \quad (4)$$

where  $\mu_s$  denotes the structural viscosity.

We assume that the pressure of the mixture is an explicit function of the mixture density, and the derivative of the pressure with respect to the density defines the modulus of compressibility  $\kappa$ , that is

$$p = p(\rho) \quad \text{and} \quad \kappa = \rho \frac{dp}{d\rho} \text{ [Pa]}. \quad (5)$$

The unknown functions in our model are density  $\rho$  and velocity  $v$ . Let us insert these functions into Eqs. (3) and (4) in the form of the sums of the average values of these functions in the equilibrium state and the fluctuations from the equilibrium due to ultrasonic wave disturbance, that is

$$\rho(x, t) = \bar{\rho} + \rho', \quad \bar{\rho} = \text{const}, \quad |\rho'| \ll \bar{\rho}, \quad (6a)$$

$$v(x, t) = \bar{v} + v', \quad \bar{v} = 0, \quad |v'| = \epsilon(0), \quad (6b)$$

where  $\rho'$  and  $v'$  denote the local and instant fluctuations of density  $\rho$  and velocity  $v$  around their equilibrium quantities  $\bar{\rho} = \text{const}$  and  $\bar{v} = 0$ .

The mass continuity equation (3) and the Navier-Stokes equation (4) expressed in terms of the fluctuations  $\rho'$  and  $v'$ , become

$$\frac{\partial \rho'}{\partial t} + \bar{\rho} \frac{\partial v'}{\partial x} = 0, \quad (7a)$$

$$\bar{\rho} \frac{\partial v'}{\partial t} = -c^2 \frac{\partial \rho'}{\partial x} + \frac{4}{3} \mu_s \frac{\partial^2 v'}{\partial x^2}, \quad (7b)$$

where  $c^2 = \kappa/\rho$  denotes the square of sound speed in the mixture. The small nonlinear terms are ignored in these equations.

Since the ultrasounds are assumed to be harmonic waves, so the same form ought to have the acoustic fluctuations. Hence, the fluctuations may be written as

$$\rho' = \rho_0 \exp[i(lx - \omega t)], \quad v' = v_0 \exp[i(lx - \omega t)], \quad (8)$$

where  $l = l_r + il_i$ ,  $l_r$  denotes the wave number,  $l_i$  is the attenuation coefficient,  $i = \sqrt{-1}$  is the imaginary number, and  $\omega$  is the angular frequency of the harmonic wave.

Substituting Eqs. (8) into Eqs. (7a) and (7b), we get

$$\rho_0 [-i\omega] + v_0 [\bar{\rho} il] = 0, \quad (9a)$$

$$\rho_0 [c^2 il] + v_0 [-\bar{\rho} i\omega - \frac{4}{3} \mu_s (il)^2] = 0. \quad (9b)$$

Equations (9a) and (9b) allow us to calculate the phase velocity and the attenuation coefficient as a function of angular frequency  $\omega$  of the ultrasonic waves, and the physical properties of the mixture represented by the structural viscosity  $\mu_s$  and the modulus of compressibility  $\kappa$  of the suspension. These equations create a homogeneous system, which may have a non-trivial solution only if the main determinant of the system creating from the terms standing at the amplitudes  $\rho_0$  and  $v_0$  is equal zero. Equating this determinant to zero, we have

$$\frac{V}{c} = \sqrt{\frac{2(1 + k^2)}{k(k + \sqrt{1 + k^2})}}, \quad (10a)$$

$$\frac{l_i}{l_r} = \frac{1}{k + \sqrt{1 + k^2}}, \quad (10b)$$

where  $V = \omega/l_r$  is the phase velocity,  $l_i$  is the attenuation coefficient, and  $\kappa$  may be written as

$$k = \frac{3}{4} \frac{\kappa}{\mu_s \omega}. \quad (11)$$

In this paper, the angular frequency  $\omega$  is a given quantity, the modulus of compressibility  $\kappa$  is to be estimated experimentally for a given suspension, and the structural viscosity  $\mu_s$  is a function of the viscosity  $\mu$  of the solvent and the volume fraction  $\phi$ . A respective relation will be derived in the Section 3.

### 3. Structural viscosity of suspensions

The motivation to study the problem of viscosity in suspensions and derivation of the relation concerning the influence of the volume fraction of solid particles on the overall viscosity is the desire to explain the experimentally observed fact that the existence of solid particles in fluid raises significantly the resistance of the two-phase medium against deformation. Each particle contributes to the local resistance of motion due to the viscous drag force. The viscosity of suspension can be assumed to be shear-rate dependent, owing to rouleaux formation of the suspension region in the presence of ultrasonic waves. The compressive action of waves on the medium has a reversible character and does not affect the viscosity.

In this paper, to derive the formula for the structural viscosity of a suspension, the solid particles are assumed to be of the form of small cylinders parallel to each other,

each of them being placed in the center of an imagined cell of a fictitious space lattice made of geometrically identical cells (Fig. 2).

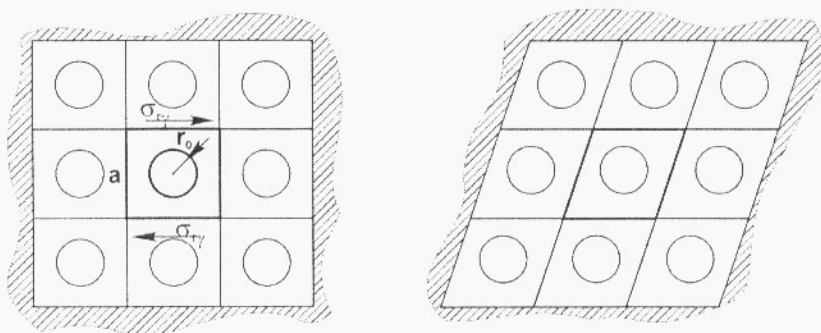


Fig. 2. Fictitious space lattice of liquid cells with solid particles in the center.

The shear-rate dependence of viscosity suggests starting the considerations from the analysis of rotational motion of liquid around a solid particle in a suspension. Let us first consider how the angular velocity depends on the radius of the particle. To this aim, it is enough to analyze the motion of viscous liquid between two rotating cylinders, Fig. 3.

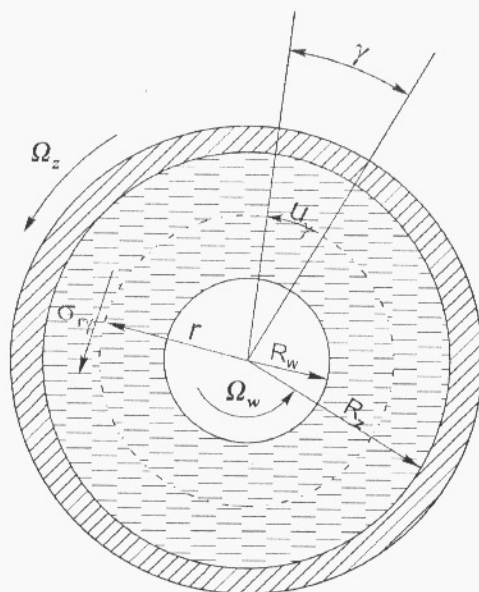


Fig. 3. Viscous liquid between two rotating cylinders.

The second Newton's law yields, for angular motion forced by rotational moment  $M$  with respect to a cylinder of radius  $r$  having the mass moment of inertia equal to  $I = mr^2/2$

$$M = \frac{d}{dt}(I\Omega) = \frac{d}{dt}\left(\frac{1}{2}mr^2\Omega\right). \quad (12)$$

This law allows us to state that the angular velocity  $\Omega$  is inversely proportional to the second power of radius, that is

$$\Omega = \frac{1}{r^2} \left[ \frac{2}{m} \left( \int_0^1 M dt + C_0 \right) \right] = \frac{C_1}{r^2} + C_2, \quad (13)$$

where  $C_1$  and  $C_2$  are constants, which are to be determined from boundary conditions. The angular velocity for viscous liquid between two rotating cylinders (Fig. 3), after determining of constants from the respective boundary conditions:  $\Omega|_{r=R_w} = \Omega_w$  and  $\Omega|_{r=R_z} = \Omega_z$ , is of the form

$$\Omega = \left[ \frac{R_w^2 R_z^2}{r^2} \frac{\Omega_w - \Omega_z}{R_z^2 - R_w^2} + \frac{\Omega_z R_z^2 - \Omega_w R_w^2}{R_z^2 - R_w^2} \right]. \quad (14)$$

In laminar angular motion, the liquid between rotating cylinders may be thought as a number of cylindrical layers rotating with different both angular and linear velocities. The shear stress between cylindrical layers is expressed by Newton's law of the form

$$\sigma_{r\gamma} = 2\mu\dot{\epsilon}_{r\gamma} \quad (15)$$

where  $(r, \gamma)$  are the spherical coordinates of a point lying on a cross-section of a cylinder, the cross-section being perpendicular to the cylinder axis (Fig. 3).

The strain  $\epsilon_{r\gamma}$  in cylindrical coordinates is

$$\epsilon_{r\gamma} = \frac{1}{2} \left( \frac{1}{2} \frac{\partial u_r}{\partial \gamma} + \frac{\partial u_\gamma}{\partial r} - \frac{u_\gamma}{r} \right) = \frac{1}{2} r \frac{\partial}{\partial r} \left( \frac{u_\gamma}{r} \right) = \frac{1}{2} r \frac{\partial \gamma}{\partial r}, \quad (16)$$

where it was substituted  $u_r = 0$  and  $\gamma = u_\gamma/r$ .

Substituting Eq. (16) into (15) and using (14), we get

$$\sigma_{r\gamma} = \mu r \frac{\partial \Omega}{\partial r} = 2\mu \frac{1}{r^2} R_w^2 R_z^2 \frac{\Omega_z - \Omega_w}{R_z^2 - R_w^2} \quad \text{where} \quad \Omega = \dot{\gamma}. \quad (17)$$



This relation will be useful in the determination of the viscous drag force acting on the solid particles in suspensions.

As the viscosity of suspension is assumed to be shear-rate dependent, let us consider the simple shearing of a separated cell of square cross-section (Fig. 2) with cylindrically shaped solid particle in the center. The shearing is induced by the acoustic field of the linearly polarized plane ultrasonic wave under consideration. Let the side of the square be much greater than the radius of the cylindrical particle, that is  $a \gg r_0$  (Fig. 4).

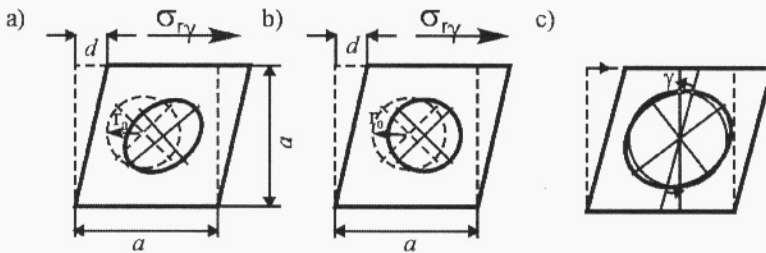


Fig. 4. Simple shearing of a separated liquid cell with cylindrical particle: a) conceived cylinder, b) rigid solid cylinder, c) comparison of the contours

Adapting the formula (17) to the geometry of Fig. 4, we take:  $R_z = a/2$ ,  $R_w = r_0$ , and  $\Omega_z = 0$ . The shear stress on the surface of the solid particle becomes

$$\sigma_{r\gamma}|_{r=r_0} = 2 \mu a^2 \frac{\Omega_w}{a^2 - 4r_0^2}. \quad (18)$$

The liquid particles adjoining the surface of the conceived cylinder displaced themselves under the action of shear forces and induce a cylinder rotation of angle  $\gamma$ . The liquid particles adjoining the solid cylinder experience only displacements, and their rotation can be consider as negligible small in comparison to those of the particles adjoining to the conceive cylinder, because of rotational inertia of the solid. We conclude then, that the solid particles involve a rheostatic braking of the liquid rotational motion with respect to solid, which would be if there were no solid particles. In order to determine the local resistance to this motion, it is necessary to calculate the power that is needed to oppose this motion with respect to solid. To this aim one needs to turn back the liquid particles on the surface of the conceived cylinder to the position of those on the solid cylinder, that this rotation of angle  $\gamma = d/a$ . The value of this power is

$$L_0 = 2\pi r_0 \sigma_{r\gamma}|_{r=r_0} \cdot \Omega_w r_0 = 4\pi \mu r_0^2 a^2 \frac{\Omega_w^2}{a^2 - 4r_0^2} \quad \text{where } \Omega_w = \dot{\gamma} \quad (19)$$

The power used for shape deformation of the liquid cell as a whole is equal to

$$L_a = 2\mu\dot{\gamma}a \cdot \dot{\gamma}a = 2\mu a^2 \dot{\gamma}^2. \quad (20)$$

Total power necessary to deformation of the liquid cell containing a solid particle is the sum

$$L = L_0 + L_a = 2a^2 \dot{\gamma}^2 \mu \left[ 1 + \frac{2\pi r_0^2}{a^2 - 4r_0^2} \right] = 2a^2 \dot{\gamma}^2 \mu_s, \quad (21)$$

where

$$\mu_s = \mu \left[ 1 + \frac{2\pi\phi}{\pi - 4\phi} \right] \quad \text{with} \quad \phi = \frac{\pi r^2}{a^2}. \quad (22)$$

Formula (22) presents the relation between structural viscosity  $\mu_s$  and the volume fraction  $\phi$ . It is seen that the structural viscosity is in general greater than the viscosity of pure solvent, and they are equal to each other only when  $\phi = 0$ . This formula shows that the structural viscosity tend to infinity when  $\phi \rightarrow \pi/4$ , what may be interpreted that for  $\phi \geq \pi/4 = 0.785$  the medium has to be no more consider as having liquefied consistency.

In the case of solid particles shaped different than cylinders, a correction factor  $\chi$  ought to be introduced to the formula (22). We suggest the general formula for structural viscosity as follows:

$$\mu_s = \mu \left[ 1 + \frac{\chi\phi}{1 - \chi\phi} \right] = \mu [1 + \chi\phi + (\chi\phi)^2 + (\chi\phi)^3 + \dots]. \quad (23)$$

The series form on the right hand side follows from the Maclaurine development.

The authors measured experimentally the viscosity of the suspension made of peat and water for a number of volume fractions. The measurements were carried out with the help of rheometer type RotoVisco 1 produced by HAAKE at the room temperatures (c.a. 20°C). The theoretical curve, constructed with the help of formula (23), and the experimental data are presented in Figure 5.

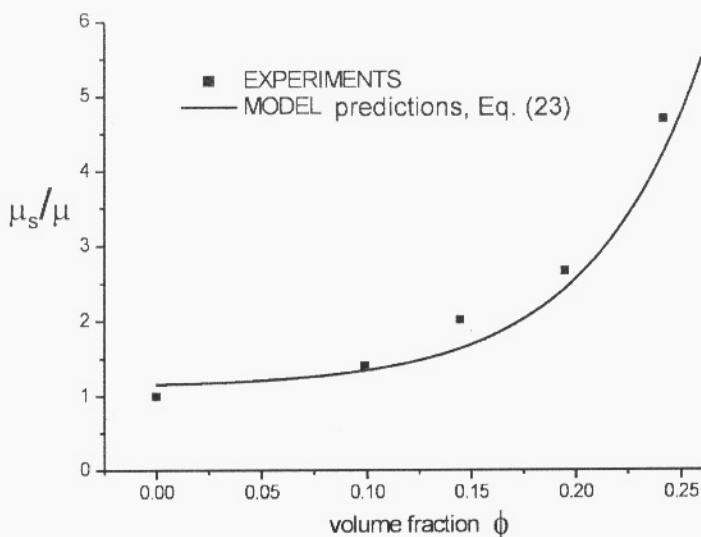


Fig. 5. Structural viscosity versus volume fraction of solid particles.

One states good agreement of the theoretical predictions with the experimental data. A similar shape of theoretical and experimental curves for structural viscosity as a function of volume fraction for sterically stabilized silica spheres suspended in cyclohexane was presented by BERGSTÖM (1994).

#### 4. Experimental determination of the phase velocity and the coefficient of attenuation for ultrasonic waves

The experiment were carried out for suspensions of peat and water of various volume fractions. The ultrasonic measurements were performed in a special tub enabling measurement *in situ*, that is, the ultrasonic heads were directly immersed in the suspension that filled up the tub. The phase velocity and the coefficient of attenuation of ultrasonic waves in the suspensions of different peat concentration were measured with the help of ultrasonic tester type UMT-01-UNIPAN. The scheme of equipment is presented in Figure 6.

The concentration of the suspensions varies from 0 to 16%. The homogeneity of the suspensions was held through continuous mixing with the help of magnetic stirrer. Because of direct contact of the ultrasonic heads with the suspension, there was no error following from the transmission of waves through the column walls (see Fig. 1).

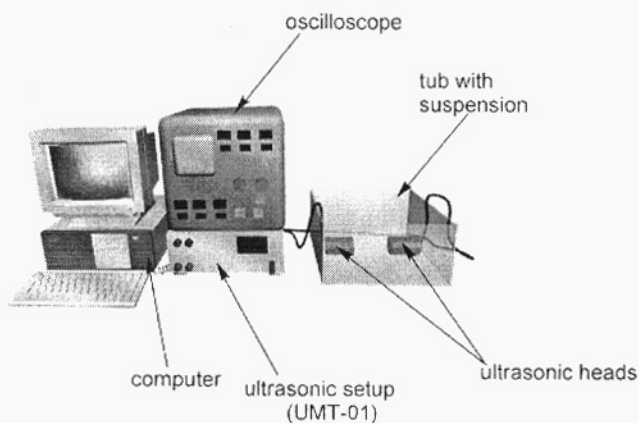


Fig.6. Ultrasonic equipment.

Figure 7 presents the relation between the velocity  $V$  of the ultrasonic wave referred to the sound speed  $c$  and the volume concentration  $\phi$  of the solid particles for dilute suspensions. The experimental results were compared with theoretical ones obtained on the basis of equation (10a) with included expressions (11) and (23). We stated a satisfactory agreement of the experimental and theoretical results.

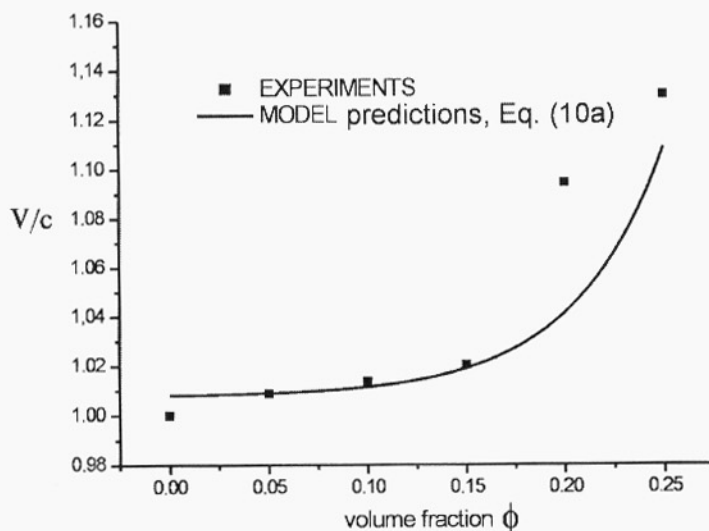


Fig. 7. Dependence between the normalized phase velocity of ultrasonic wave and the volume fraction of the solid particles.

Figure 8 presents the coefficient of attenuation  $l_i$  of ultrasonic wave referred to the wave number  $l_r$  as a function of volume fraction  $\phi$  for the angular frequency  $\omega = 600$  kHz.

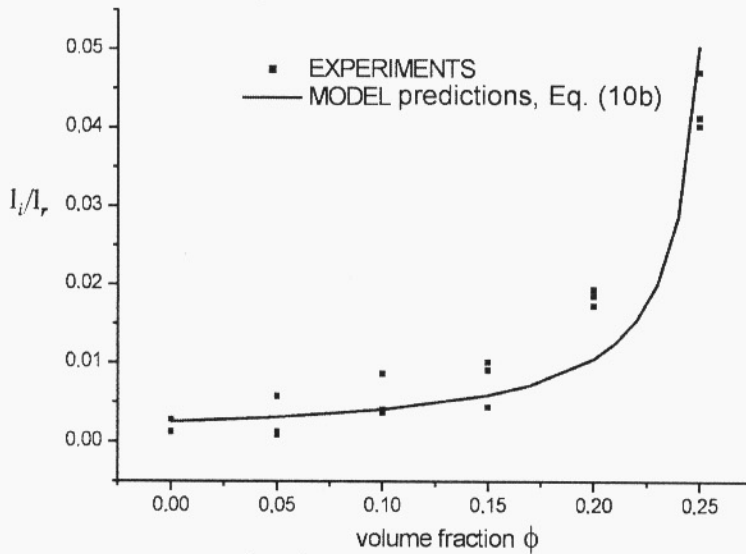


Fig. 8. Normalized attenuation coefficient versus volume fraction of the solid particles.

### 5. Final remarks

Our preliminary experiments show that it is possible to use the ultrasonic method for determination of densities of suspensions and sediments. Some difficulties, which arose during our experimental studies concerned mainly of a proper choice of the power and frequency of the ultrasonic waves for the given suspension. If one chooses unsuitable frequency or power, the measurement of the density may be not possible, because of dispersion and damping of the waves. The emitted wave may not arrive to the receiving head.

As it results from equation (11), the attenuation coefficient  $l_i$  depends on both the suspension concentration  $\phi$  and the wave frequency  $\omega$ . The wave frequency should be so chosen to obtain a minimal attenuation coefficient. The confinement of the experimental studies to suspensions of small concentration was limited by the theoretical model, which was elaborated for diluted suspensions. The studies, both theoretical and experimental, for higher concentrations will be continued, based on the author's earlier concept of consolidation theory (see e.g. DERSKI and KOWALSKI (1979), KOWALSKI (1983)).

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