A New Technique for the Investigation of Sedimentation Under Gravity

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A new, non-destructive contactless technique: Positron Annihilation Gamma Probe for measuring the density profiles of highly-concentrated sedimenting systems using narrow-beam monoenergetic positron annihilation gamma-quanta is suggested.

The investigation of sedimentation processes in highly-concentrated disperse systems is one of the most important problems in modern colloid chemistry. This is associated both with the necessity of developing fundamental aspects of the problem and with the great variety of diverse heterogeneous chemical processes that are accompanied by sedimentation phenomena.[1]

Up to now, the absence of reliable techniques sufficiently sensitive to structural changes has been a substantial obstacle in the way of investigating sedimentation processes in concentrated systems. Gamma ray attenuation metering is an established method with which to determine the density profiles of various substances.[2,3] In positron annihilation spectroscopy the original methods of production, forming and detection of narrow monochromatic (511 keV) annihilation gamma quanta beams has been developed.[4] Thus, Positron Annihilation Gamma Probe (PAGP) appears to be a very convenient technique for density profiles measurements.

For a two-phase system of the type 'dispersion medium – disperse phase', when using the gamma-ray technique for the determination of density in a modification of the geometry of a narrow beam the attenuation of the initial beam when passing through the substance is described by the exponential Beer–Lambert law. The counting rate of gamma quanta in a detector can be expressed as equation (1),

$$I = I_b + A\exp\left[-\rho d\left(P_1\frac{\mu_1}{\rho_1} + P_2\frac{\mu_2}{\rho_2}\right)\right] \quad (1)$$

where ρ_1, P_1 and μ_1 are the mass density, the weight fraction and the extinction coefficient of the dispersion medium, respectively, while ρ_2, P_2 and μ_2 are the corresponding values for disperse phase; ρ is the mass density of the system; d is the length of the substance layer; A is the coefficient taking into account the intensity of the primary gamma quanta beam and attenuation of the beam by the container material; and I_b is the background counting rate in the detector.

To determine the weight or volume fraction of one of the components (for example, of the disperse phase) one should know the extinction coefficients for each compound of the system and of the vessel material at the point where the gamma beam has passed through. This usually seems to be impossible even for a two-phase system. In Bergstrom's papers[2,3] this problem was solved by measuring the calibration curves, thereby obtaining an empirical dependence of the magnitude of attenuation as a function of volume fraction of the disperse phase in specially prepared model two-phase systems. The calibration curve thus obtained is further used for the density profile calculations. However, the systems utilized for carrying out such a calibration should possess an exceptional colloid stability, resistance in relation to sedimentation and have a strictly constant density over the height of the container which is used in further measurements. This imposes rigorous limitations upon a variety of objects being measured. Bergstrom's method[2,3] is unsuitable for systems sedimenting at a noticeable rate under gravity.

The problem can be solved by means of measuring with one and the same position of the gamma quanta beam the count rate in the detector for the empty container I_a (when effective density may be considered equal to zero $\rho = 0$), then for the container filled up with only a dispersion medium I_m ($\rho = \rho_1$), and with only a dispersed phase I_f (the volume density $\rho = \rho_f$). By placing a thick (10 cm) lead brick between gamma source and detector

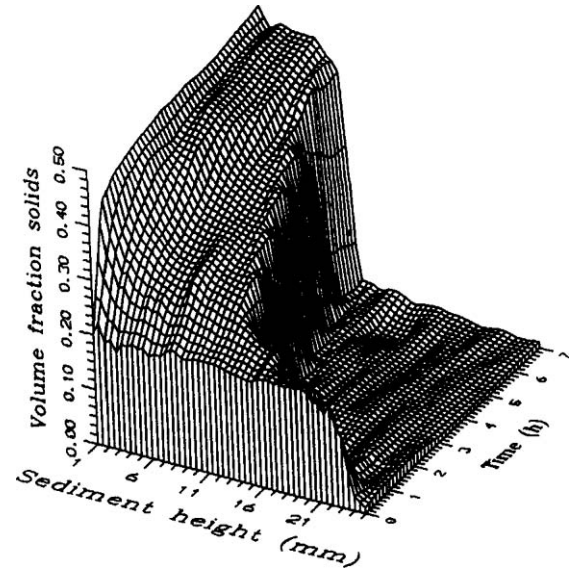

Fig. 1 Evolution of volume fraction solid profiles (volume fraction of disperse phase V_s *versus* sediment height) for an iron oxide water-glycerine highly-concentrated (50% mass) suspension.

($\rho \simeq \infty$) one can measure the background count rate I_b. Substituting in each case these values into equation (1) and solving the set of equations one can calculate the dependence of the volume fraction of the disperse phase value V_s in a layer of the system being probed under the count rate in the detector I, equation (2),

$$V_s = \ln\left(\frac{I - I_b}{I_m - I_b}\right) \Big/ \left[\ln\left(\frac{I_a - I_b}{I_m - I_b}\right) + \frac{\rho_2}{\rho_f}\ln\left(\frac{I_f - I_b}{I_a - I_b}\right)\right] \quad (2)$$

Thus, measuring the count rate in the detector, it is possible to obtain the distribution of the volume fraction of the disperse phase over the height (volume fraction solids profile) which is one of the most important characteristics of the sedimenting system.

In Fig. 1 are presented as an example the results of an investigation of the settling process of γ–Fe_2O_3 powder in a water–glycerine medium. The powder particle size is *ca.* 3–4 μm, the mass concentration of the dispersed phase *ca.* 50%. The disperse medium was prepared by mixing 200 cm^3 of glycerine with 50 cm^3 of water. A container of rectangular shape made of acrylic plastic was used as a vessel, with wall thickness 5 mm and internal dimension $d = 70$ m. The positron source was an 22 Na isotope of activity *ca.* 5 mCi; the width of an annihilation gamma quanta beam, emitted from an aluminium target, was 0.8 mm. Scanning with an annihilation gamma probe over the height of the system was effected by shifting the container vertically. The time of a single measurement was 30 s, the measurements being carried out in steps of 1 mm over the height. The accuracy of determination of effective mass density was equal to 0.2% with the aforesaid measurement parameters.

In Fig. 1 is shown how the volume fraction of the dispersed phase V_s changes over the height of a container in the system under investigation during the sedimentation process. Prior to beginning the measurements the system was thoroughly mixed

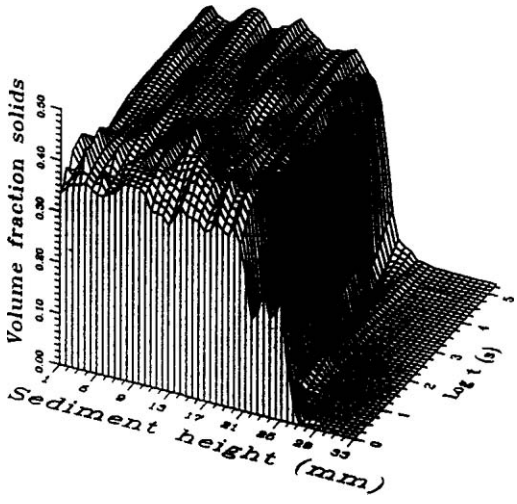

Fig. 2 Formation of different density periodical layers in a sediment of highly-concentrated (75% mass) disperse Zn – water glass system.

to attain homogeneity. The settling time was in the range of 0.5–7 h. The formation of a distinct jump in density is clearly visible, corresponding to the interface between a dense sediment and a disperse medium. As settling proceeds, the jump in density near the interface shifts to the vessel bottom, and its position is practically stabilized when the settling time exceeds 7 h. The volume fraction of dispersed phase profile in a sediment layer has a complex character. At the initial settling stages the density of the sediment increased monotonously with depth down to the vessel's bottom. Beginning with a settling time of 3 h and greater, attention is drawn to the formation and increase of the local density minimum at a height of *ca.* 2 mm in the sediment layer. This phenomenon also seems to be observed in Bergstrom's paper,[2] but the author has probably not paid attention to the peculiarities of density profiles, exhibiting a weakly pronounced character.

The PAGP technique reveals delicate structural details in the sediment density profiles. In Fig. 2 a very interesting phenomenon of different density periodical layer formation in a sediment of highly-concentrated disperse Zn – water glass system is shown. The mass concentration of the solid phase was 75%, Zn particle mean size 10 μm, and sedimentation took place in a glass cylindrical vessel of 32 mm diameter. A similar flaky sediment structure was also discovered in carbon black – water highly concentrated suspensions.

In conclusion, the PAGP technique may be applied to study sedimentation processes in highly-concentrated disperse systems, where the application of conventional colloid chemistry techniques is rendered extremely difficult.

References

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