

An electrical conductivity probe method for measuring the local solid holdup in a slurry system

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Abstract

An electrical conductivity probe method for measuring the local solid holdup in a liquid–solid or gas–liquid–solid slurry system was developed. The relationship between the conductivity of the liquid–solid mixture and the local solid holdup was described by the electric field theory. From this relationship, the local solid holdup can be determined from the measured conductivity of the liquid–solid mixture. Validation was carried out for several cases. The experimental results showed that the conductivity probe method developed in this work was convenient for use and had a good accuracy in a wide range of solid holdup in the liquid–solid system. By adding sieves at the front of the probe to eliminate the effect of gas bubbles, the modified probe was used to measure the local solid holdup in a gas–liquid–solid system and good results were obtained.

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Keywords: Local solid holdup; Electrical conductivity probe; Gas–liquid–solid; Measuring method

1. Introduction

Liquid–solid and gas–liquid–solid multiphase reactors are promising devices in chemical, biochemical and environmental engineering operations. Extensive studies on hydrodynamics and mass and heat transfer have been reported in the past years [1,2]. In liquid–solid and gas–liquid–solid reactors, the solid particles are usually catalyst, thus increasing solid holdup can increase the reaction rate and the reactor efficiency. However, the apparent viscosity of the liquid–solid suspension increases with the solid holdup, and the non-uniform profiles of the solid holdup will become marked in the range of high solid holdup. The solid holdup and its profiles have great influence on the hydrodynamics, mass-transfer behavior and reactor efficiency. In three-phase fluidized bed reactor with particle size of millimeters, the solid holdup has clear non-uniform axial and radial profiles. Comprehensive studies were reported concerning on the non-uniform profile of solid holdup, its impact on hydrodynamics and the mechanism of forming such non-uniformity in a three-phase fluidized bed [3,4]. In a slurry system

where the particles were fine (10–100 μm), most works in the literature showed that the solid holdup was almost uniform [5]. However, when the solid holdup was further increased, the non-uniform profiles of solid holdup were also found in a slurry system [6]. Thus, it is important to measure the local solid holdup in the three-phase fluidized bed and slurry reactors.

Many non-intrusive or intrusive measurement techniques have been developed in the last decade to measure the local solid holdup or gas holdup in the two- or three-phase systems. Gandhi et al. [5] studied the solid distribution in a slurry bubble column using a sampling probe. This method was also used by Kuramoto et al. [7] in a liquid–solid fluidized bed reactor. The sampling method is simple in principle, but suffers from many uncertainties that affect the sampling accuracy and the data cannot be obtained on-line. Furthermore, the sampling method cannot be used at high solid holdup conditions. Wenge et al. [8] used the dynamic gas disengagement (DGD) method to measure the gas and solid holdups in the three-phase systems. The DGD method has some limitations in practical applications. The severe fluctuations in the measured pressure cause remarkable error. Further, it cannot be used to measure the local phase holdups. Warsito et al. [9] measured the radial profile of the solid holdup in a slurry bubble column by an ultrasonic probe,

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Nomenclature

a_1	radius of fine particles (mm)
a_2	radius of the measuring sphere (mm)
d_p	fine particle size (μm)
k_1	resistivity of fine particles ($\text{cm}/\mu\text{S}$)
k_2	resistivity of liquid between electrodes ($\text{cm}/\mu\text{S}$)
K	resistivity of pseudo-homogenous phase ($\text{cm}/\mu\text{S}$)
m_s	weight of fine particles added (kg)
r	radial distance (mm)
U_g	superficial gas velocity (m/s)
V_1	liquid volume (m^3)
V_1	the electrical potential function in fine particles
V_2	the electrical potential function in the liquid between electrodes

Greek symbols

ε_s	solid holdup
$\varepsilon_{s,r}$	real solid holdup
$\varepsilon_{s,m}$	measured solid holdup
γ	conductivity of mixture ($\mu\text{S}/\text{cm}$)
γ_0	conductivity of liquid ($\mu\text{S}/\text{cm}$)
ρ_s	material density of fine particles (kg m^{-3})

and the results needed modification by a correction factor to reduce the effect of insertion of ultrasonic probe. Warsito et al. [4] also used the ultrasonic computed tomography (UCT) to measure the cross-sectional profile of the gas and solid holdups in a slurry bubble column. George et al. [10] expatiated on gamma-densitometry tomography (GDT) method and electrical-impedance tomography (EIT) method to measure the three-phase profiles in a vertical flow. The outstanding characters of UCT, GDT and EIT are their non-intrusive operation, however, these non-intrusive methods are much more expensive and the temporal and spatial resolutions are still to be improved.

Electrical conductivity technique is another important method for measuring the solid holdup. The principle was compendiously mentioned in the masterpiece of Maxwell [11], and no detailed discussion was available. Most succeeding researchers focused on the use of the electrical conductivity probe without an in-depth discussion of the method itself [12–16]. Further, only the averaged solid holdup was measured in most of these works.

This work aims to give a detailed discussion on the theoretical basis of the electrical conductivity probe method and propose a probe method for measuring the local solid holdup in both liquid–solid and gas–liquid–solid systems. A clear deduction of the influence of solid particles on the electric field, from one particle to multi-particles, was presented. The assumptions during the deduction and their effects on the application of the probe were analyzed to give guidance for the appropriate use of this method. The experimental results showed that the particle shape had remarkable effects, but the effect of the solid holdup was neglectable. Actually, the probe still had a good accuracy

in a liquid–solid system at solid holdup up to 40%. Based on this analysis, we found that a larger error with the conductivity probe used at relatively high gas holdup was mainly due to bubble deformation, not caused by the high gas holdup as reported by Uribe-Salas et al. [14].

2. Mathematical model

2.1. Single particle case

We first consider the case with a single fine spherical particle of radius a_1 between the two metal electrodes, as shown in Fig. 1. The space between the electrodes is called as the measuring volume. The resistivities of the particle and the liquid between the two electrodes are k_1 and k_2 , respectively. The electrical potential functions (EPF) inside and outside the particle are denoted as V_1 and V_2 , respectively. Because there is no electrical source in the measuring volume, the EPFs satisfy the Laplace's equation:

$$\nabla^2 V_1 = 0, \quad \nabla^2 V_2 = 0 \quad (1)$$

In a spherical coordinate as shown in Fig. 2, the boundary conditions are:

- $r=0$, V_1 is finite.
- $r=a_1$, $V_1 = V_2$.
- $r/a_1 \rightarrow \infty$, $V_2 = -E_0 Z = -E_0 r \cos \theta$.

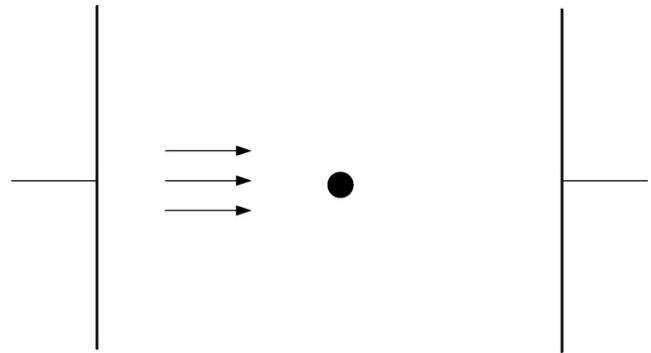


Fig. 1. Schematic representation of the electrode and the single particle.

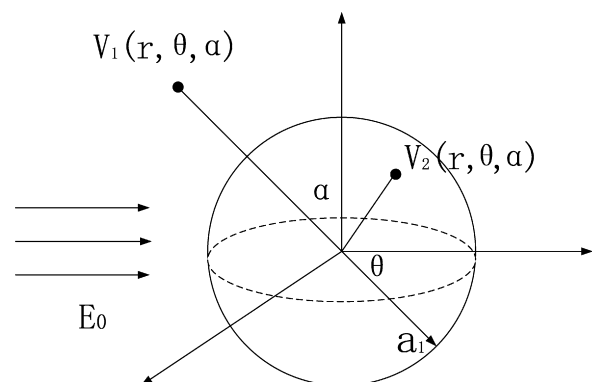
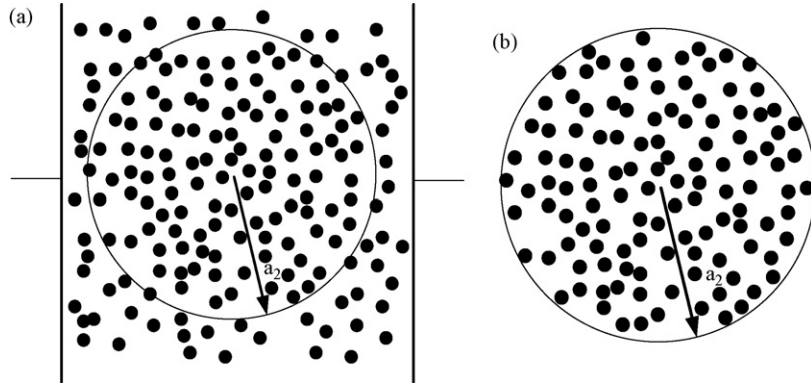


Fig. 2. Schematic representation of the 3D sphere coordinate (single particle).

Fig. 3. Schematic representation of the measuring system (n particles).

The EPFs do not change with the azimuth angle in the spherical coordinate, so the general solution of the Laplace's equation is (see Appendix A):

$$V_1 = \sum_{n=0}^{\infty} \left(A_n r^n + \frac{B_n}{r^{n+1}} \right) P_n(\cos \theta) \quad (2)$$

$$V_2 = \sum_{n=0}^{\infty} \left(C_n r^n + \frac{D_n}{r^{n+1}} \right) P_n(\cos \theta) \quad (3)$$

where $P_n(\cos \theta)$ is Legendre polynomial, $P_0(\cos \theta) = 1$ and $P_1(\cos \theta) = \cos \theta$. The details are given in Appendix A.

With the consideration of the boundary conditions, the following results are obtained:

- Because $r \rightarrow \infty$, $V_2 = -E_0 r \cos \theta = -E_0 r P_1(\cos \theta)$, V_2 has only the “ $n = 1$ ” item and $C_1 = -E_0$.
- Because $V_1 = V_2$ at $r = a_1$, and V_2 has only the “ $n = 1$ ” item, V_1 has only the “ $n = 1$ ” item as well.
- Because V_1 is finite at $r = 0$, $B_1 = 0$.

Substitution of the above results into the general solution of the Laplace's equation yields:

$$V_1 = A_1 r \cos \theta \quad (4)$$

$$V_2 = \left(-E_0 r + \frac{D_1}{r^2} \right) \cos \theta \quad (5)$$

At $r = a_1$, V_1 and V_2 satisfy the following relationship:

$$V_1 = V_2, \quad \frac{1}{k_1} \frac{dV_1}{dr} = \frac{1}{k_2} \frac{dV_2}{dr} \quad (6)$$

Substitution of Eqs. (4) and (5) into Eq. (6) reads:

$$A_1 = -E_0 + \frac{D_1}{a_1^3} = -\frac{k_1}{k_2} \left(E_0 + \frac{2D_1}{a_1^3} \right) \quad (7)$$

The coefficients A_1 and D_1 are:

$$A_1 = -\frac{3k_1}{k_2 + 2k_1} E_0, \quad D_1 = \frac{k_2 - k_1}{k_2 + 2k_1} E_0 a_1^3 \quad (8)$$

The final solutions of V_1 and V_2 are:

$$V_1 = -\frac{3k_1}{k_2 + 2k_1} E_0 r \cos \theta, \quad V_2 = -E_0 \left(r + \frac{k_1 - k_2}{2k_1 + k_2} \frac{a_1^3}{r^2} \right) \cos \theta \quad (9)$$

2.2. Multiple particles case

The case of multiple particles considered here satisfies the condition that the measuring volume is small enough compared with the reactor and large enough compared with the solid particles in the slurry system. In such a case, the following assumptions are made to deduce the relationship between the electrical conductivity of the liquid–solid slurry and the solid holdup.

- (1) The particles are spherical and have the same radius a_1 .
- (2) The particle size is small enough compared with the measuring space so that their interferences on the electrical field are mutually independent.
- (3) The particles are homogeneously suspended in the measuring volume, thus the conductivity of the measuring volume in Fig. 3(a) is equal to that of the sphere volume between the two electrodes in Fig. 3(b). The radius of the sphere volume is a_2 that ensures the volume contains enough particles.

The above conditions are commonly well satisfied in a slurry system. The particles are homogeneously suspended, therefore every particle has one point-symmetry particle with respect to the center of the measuring volume. Such two point-symmetry particles have the same effect on the EPF as that of the two center-particles as shown in Fig. 4. For n particles, the effects of the particles on the electric field are mutually independent and can be determined by superposition of the effect of n center-particle particles. Thus, the EPF in the liquid with n particles in the measuring volume can be obtained from Eq. (9) as follows:

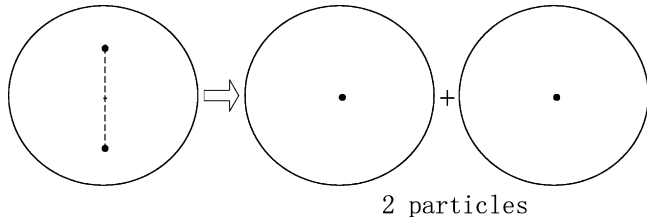


Fig. 4. Equivalence of the two point-symmetry particles to the two center-particles on EPF.

$$V_2 = -E_0 r \cos \theta - \underbrace{\left(E_0 \frac{k_1 - k_2}{2k_1 + k_2} \frac{a_1^3}{r^2} \cos \theta + E_0 \frac{k_1 - k_2}{2k_1 + k_2} \frac{a_1^3}{r^2} \cos \theta + \dots \right)}_n \quad (10)$$

The first item in the right-hand side of Eq. (10) is the origin EPF, and the other items stand for the influence of the fine particles, as shown in Fig. 5. An equivalent form of Eq. (10) is

$$V_2 = -E_0 \left(r + \frac{k_1 - k_2}{2k_1 + k_2} \frac{na_1^3}{r^2} \right) \cos \theta \quad (11)$$

The solid holdup is defined as the volume fraction of solid particles, and can be expressed as:

$$\varepsilon_s = \frac{na_1^3}{a_2^3} \quad (12)$$

Combination of Eqs. (11) and (12) yields:

$$V_2 = -E_0 \left(r + \varepsilon_s a_2^3 \frac{k_1 - k_2}{2k_1 + k_2} \frac{1}{r^2} \right) \cos \theta \quad (13)$$

Suppose the liquid–solid phase is pseudo-homogenous and its resistivity is K , this leads to $\varepsilon_s = 1$. Then the flowing equation is obtained from Eq. (13):

$$V_2 = -E_0 \left(r + a_2^3 \frac{K - k_2}{2K + k_2} \frac{1}{r^2} \right) \cos \theta \quad (14)$$

Combination of Eqs. (13) and (14) gives:

$$\varepsilon_s \frac{k_1 - k_2}{2k_1 + k_2} = \frac{K - k_2}{2K + k_2} \quad (15)$$

i.e.,

$$K = \frac{2k_1 + k_2 + \varepsilon_s(k_1 - k_2)}{2k_1 + k_2 - 2\varepsilon_s(k_1 - k_2)} k_2 \quad (16)$$

where K is the resistivity of the liquid–solid mixture, and k_2 is the resistivity of the liquid. The electrical conductivity of the liquid–solid mixture γ and the conductivity of the liquid γ_0 are:

$$\gamma_0 = \frac{1}{k_2}, \quad \gamma = \frac{1}{K} \quad (17)$$

Substituting Eq. (17) into Eq. (16) yields:

$$\frac{\gamma_0}{\gamma} = \frac{2k_1 + k_2 + \varepsilon_s(k_1 - k_2)}{2k_1 + k_2 - 2\varepsilon_s(k_1 - k_2)} \quad (18)$$

For non-electric particles, k_1 is infinite, therefore Eq. (18) is simplified as

$$\begin{aligned} \frac{\gamma_0}{\gamma} &= \frac{2k_1 + k_2 + \varepsilon_s(k_1 - k_2)}{2k_1 + k_2 - 2\varepsilon_s(k_1 - k_2)} \\ &= \frac{2 + (k_2/k_1) + \varepsilon_s(1 - (k_2/k_1))}{2 + (k_2/k_1) - 2\varepsilon_s(1 - (k_2/k_1))} = \frac{2 + \varepsilon_s}{2 - 2\varepsilon_s} \end{aligned}$$

i.e.,

$$\varepsilon_s = \frac{2 - 2\gamma/\gamma_0}{2 + \gamma/\gamma_0} \quad (19)$$

For a liquid–solid system, the liquid conductivity γ_0 is known, and the solid holdup ε_s can be determined from Eq. (19) if the conductivity of the liquid–solid slurry γ is measured.

3. Experimental

The experiments were carried out in a stirred vessel, as shown in Fig. 6. Non-electric particles (glass beads and silicon particles) and tap water were used as the solid and liquid phases, respectively. The properties of the materials used were listed in Table 1. The raw glass beads have some electrolyte adhered on their surface, which can be removed according to the experiment requirement by bathing with de-ionized water for several times. The conductivity was measured on-line by the DDSJ-308 conductivity meter. The conductivity of the pseudo-homogeneous liquid–solid slurry γ was measured after the slurry was well mixed. And then, the stir was stopped and after all glass beads had completely settled, the conductivity of the liquid γ_0 was measured in the upper region free of particles. The solid holdup $\varepsilon_{s,m}$ was determined by Eq. (19) and compared with the real solid holdup. The real solid holdup $\varepsilon_{s,r}$ and the mass m_s of particles added to the system have the following relationship:

$$\varepsilon_{s,r} = \frac{m_s/\rho_s}{m_s/\rho_s + V_1} \quad (20)$$

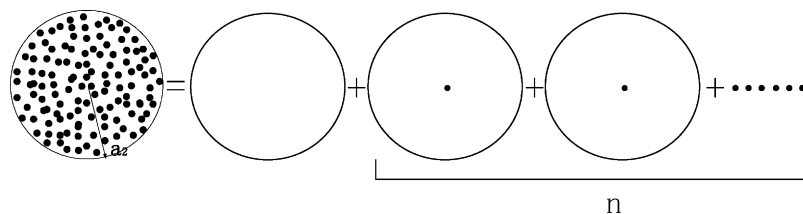


Fig. 5. Schematic representation of Eqs. (10) and (11).

Table 1
Properties of experimental materials

Material	Substance	Size, d_p (μm)	Viscosity (Pa s)	Density (kg m^{-3})
Liquid	Water	–	0.9×10^{-3}	998
Gas	Air	–	1.0×10^{-5}	1.2
Fine particle 1	Glass beads	53, 83, 117	–	2526.1
Fine particle 2	Silicon particles	57	–	2454.5



Fig. 6. Schematic representation of the experimental apparatus.

where ρ_s is the material density of the particles; V_l the water volume; m_s is the mass of added glass beads.

Because the electrode of the origin probe is bared directly in the measured system, bubbles will enter the measuring volume when using the probe to measure a gas–liquid–solid system, as shown in Fig. 7(a). The bubbles in the measuring volume remarkably decrease the conductivity and will cause marked error in

measurement of the solid holdup. One way to solve this problem is to determine the gas and solid holdups simultaneously [14]. In the approach proposed by Uribe-Salas et al. [14], the bubble size must be smaller than 2 mm so that the bubbles are approximately spherical, however, this condition cannot be satisfied in most practical systems, especially at high solid holdups. In our experiments, the probe was modified by adding sieve in the direction of coming flow, and the sieve has such holes (60 mesh in this work) that allow the pass of fine particles but hinder the pass of gas bubbles, as shown in Fig. 7(b) and (c). Experiments were carried out both in gas–liquid and liquid–solid systems with the modified probe to check the effect of the added sieve on the measurement accuracy.

4. Results and discussion

4.1. Liquid–solid system

4.1.1. Effect of electrolyte

4.1.1.1. *Glass beads with/without electrolyte adhered.* The raw glass beads have some electrolyte adhered on their surface. When using such glass beads directly, the electrical conductivity of the slurry increased with increasing solid holdup because additional electrolyte was introduced together with the glass beads, as shown in Fig. 8. However, the relative electrical conductivity γ/γ_0 decreased with increasing solid holdup, and Eq. (19) was still adequate to determine the solid holdup. Good agreement between the real and measured solid holdups was

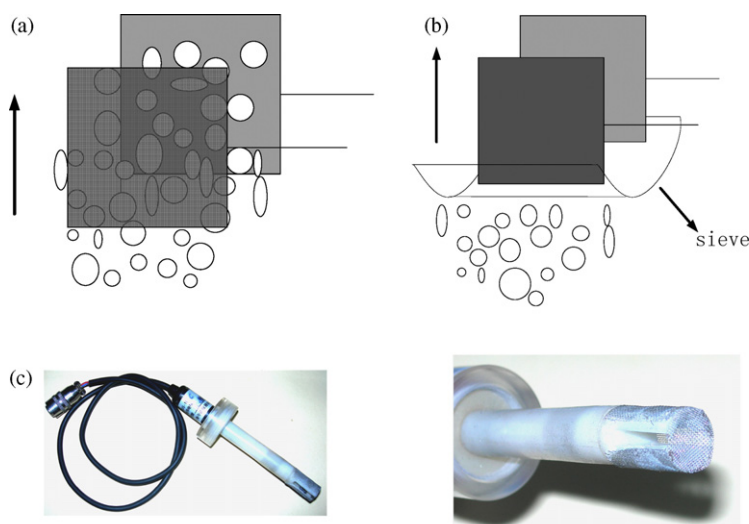


Fig. 7. The electrical conductivity probe: (a) the origin probe (gas–liquid system); (b) the modified probe (gas–liquid system); (c) the photos of the original and modified probes.

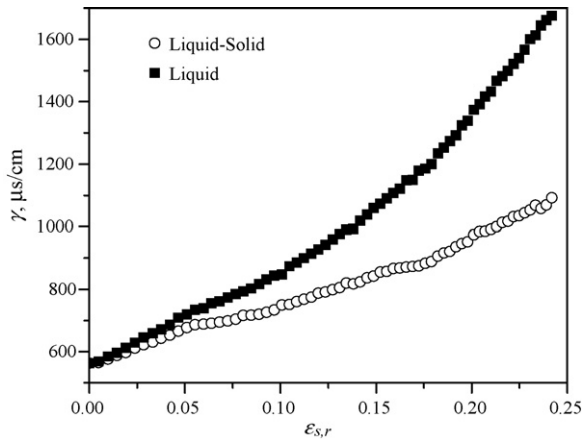


Fig. 8. Relationship between the electrical conductivity and the solid holdup with electrolyte adhered (glass beads, $d_p = 53 \mu\text{m}$).

obtained, as shown in Fig. 9. The results showed that the method still had a good accuracy even at high solid holdup up to 25%.

To further study the effect of the electrolyte adhered on surface of the particles, the glass beads were bathed with de-ionized water for several times. The change of the electrical conductivity with increasing amount of added glass beads was shown in Fig. 10. In such a case, the electrical conductivity of the liquid almost unchanged and the conductivity of the slurry increases with increasing solid holdup. The agreement between the real and measured solid holdups is satisfactory, as shown in Fig. 11.

4.1.1.2. Glass beads in the KCl solution. Similar experiments were carried out with glass beads in KCl solution to study the influence of the electrolyte concentration in the liquid. Glass beads were added to KCl solution with a conductivity of $1580 \mu\text{S}/\text{cm}$. The change of the electrical conductivity was shown in Fig. 12, and the comparison between the real and measured solid holdup was shown in Fig. 13.

The measured solid holdup was still in a good agreement with the real solid holdup, showing that the electrical concentration in the liquid has no influence on the accuracy of this

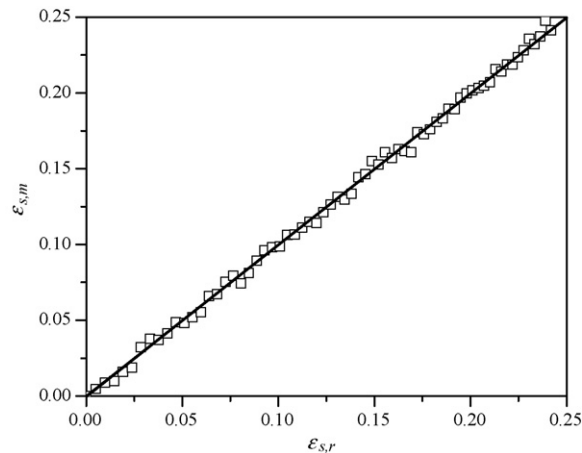


Fig. 9. Comparison between $\epsilon_{s,m}$ and $\epsilon_{s,r}$ with electrolyte adhered (glass beads, $d_p = 53 \mu\text{m}$).

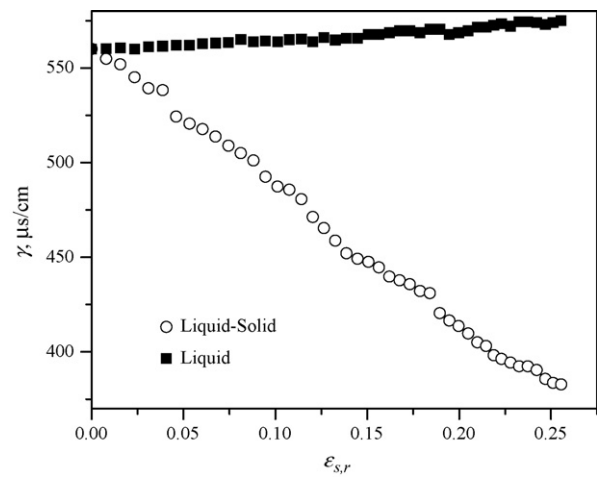


Fig. 10. Relationship between the electrical conductivity and the solid holdup without electrolyte adhered (glass beads, $d_p = 53 \mu\text{m}$).

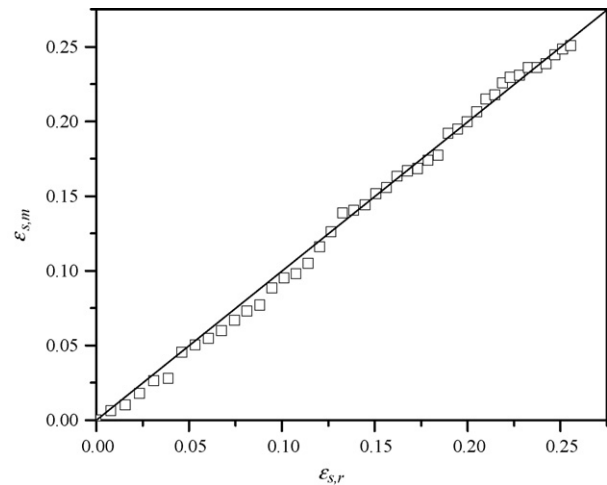


Fig. 11. Comparison between $\epsilon_{s,m}$ and $\epsilon_{s,r}$ without electrolyte adhered (glass beads, $d_p = 53 \mu\text{m}$).

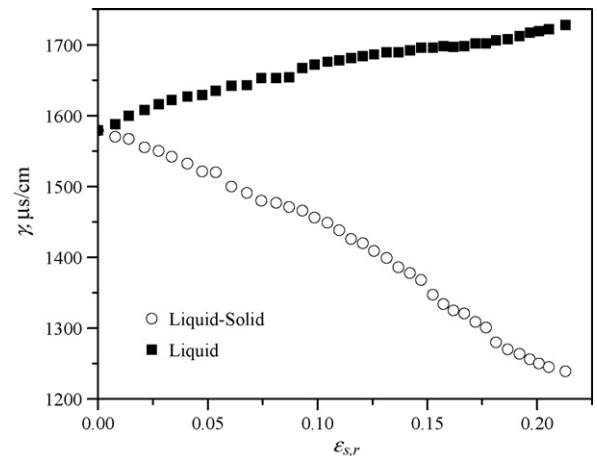


Fig. 12. Relationship between the electrical conductivity and the solid holdup in the KCl solution (glass beads, $d_p = 53 \mu\text{m}$).

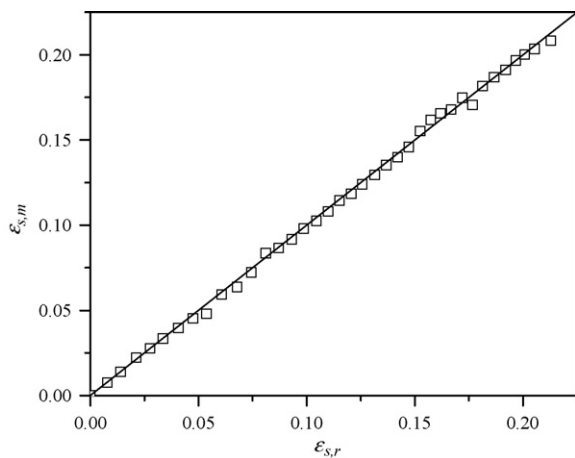


Fig. 13. Comparison between $\varepsilon_{s,m}$ and $\varepsilon_{s,r}$ in the KCl solution (glass beads, $d_p = 53 \mu\text{m}$).

measuring method. Therefore, some electrolyte can be added to increase the liquid conductivity when the conductivity of the liquid–solid system is too small, and de-ionized water can be added to decrease the liquid conductivity to meet the measuring range of the conductivity meter.

4.1.2. Effect of particle shape

Experiments were also carried out using silicon fine particles with irregular shape to study the influence of the particle shape on the method. The shape of silicon particles was measured by SEM, as shown in Fig. 14. The results showed that the particle shape had great influence on the measured solid holdup, as shown in Fig. 15. This is because Eq. (19) is deduced with the assumption that particle is spherical. Further work is needed to extend this conductivity probe method to a system with irregular particles. It should be noted that due to the remarkable influence of the particle shape, the conductivity method cannot be used to measure the gas holdup of distorted bubbles.

From the discussion above, it can be seen that Eq. (19) describe the relation between the mixture electrical conductivity and the solid holdup very well for spherical glass beads, either in tap water or in KCl solution. It is feasible to use this method to measure the local solid holdup in a liquid–solid system. In

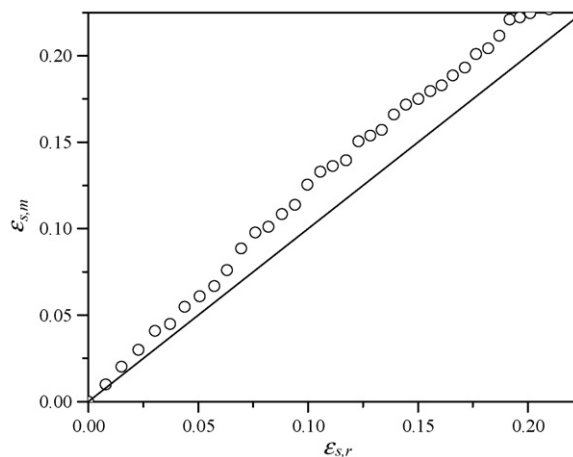


Fig. 15. Comparison between $\varepsilon_{s,m}$ and $\varepsilon_{s,r}$ with silicon fine particles.

the above experiments, glass beads with electrolyte adhered has the best accuracy ($\pm 0.8\%$). The electrolyte concentration in the liquid has no influence on the measuring accuracy, but the particle shape has a significant effect. The method still has good accuracy even at high solid holdup up to 25% or ever higher, thus can be used to measure the local solid holdup in a reactor operated at high solid concentrations.

4.2. Gas–liquid–solid system

4.2.1. Performance of the modified probe for hindering bubbles

Experiments were carried out in a gas–liquid system both with the original probe and the modified probe to check the performance of the modified probe for hindering bubbles. The results at different superficial gas velocities were shown in Fig. 16 and Table 2. The performance of the modified probe for hindering bubbles was satisfactory. Even at a superficial gas velocity of 28.4 cm/s, the decrease in the conductivity resulted from tiny bubbles will only cause an equivalent error of 2.86% solid holdup according to Eq. (19). The error can be further decreased when the superficial gas velocity is smaller. Because the electrolyte concentration in the liquid has no influence on

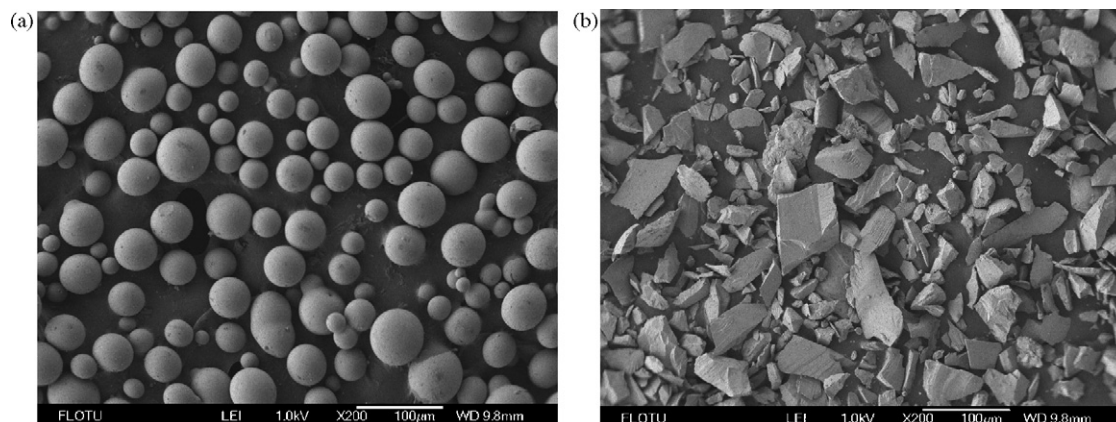


Fig. 14. The shape of fine particles: (a) glass beads; (b) silicon fine particles.

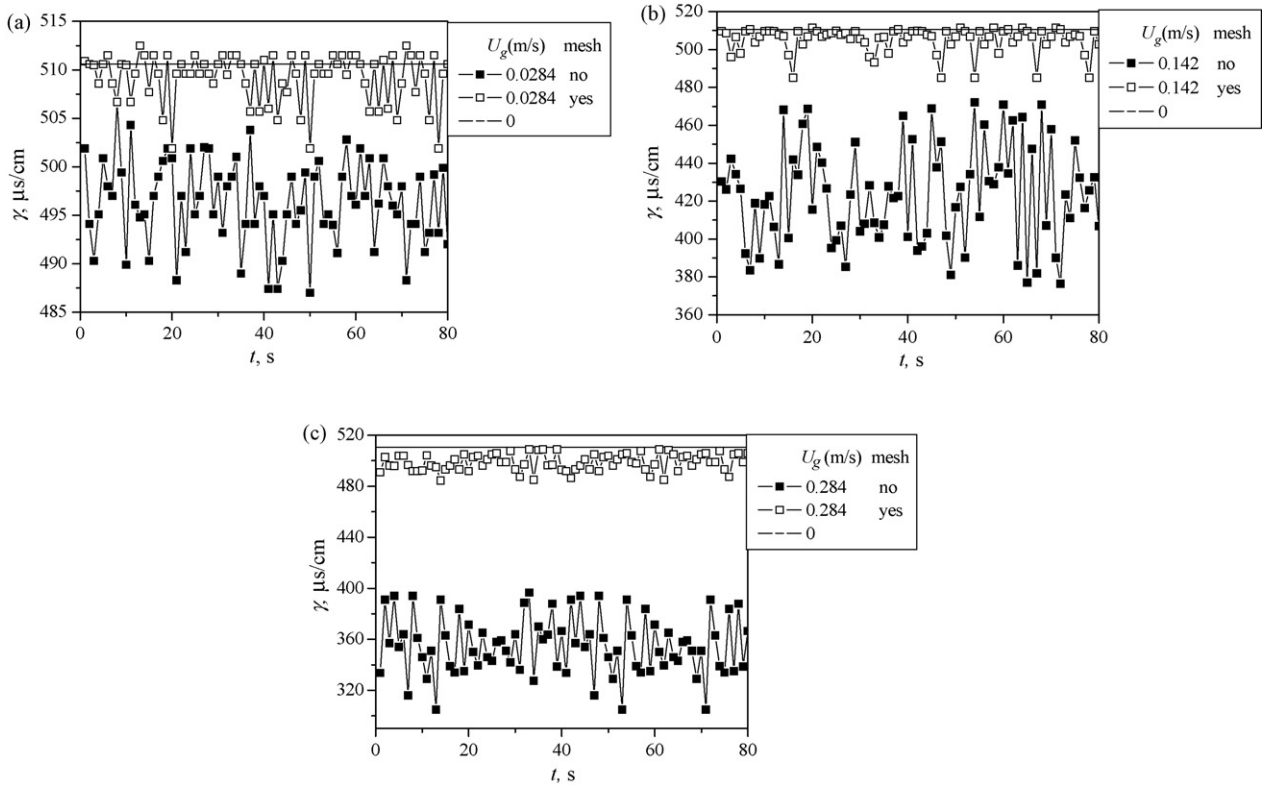


Fig. 16. Experiment results with the origin and modified probes in the gas-liquid system: (a) $U_g = 0.0284$ m/s; (b) $U_g = 0.142$ m/s; (c) $U_g = 0.284$ m/s.

the measuring accuracy, the absolute decrease of conductivity due to tiny bubbles will not change at a given superficial gas velocity, and the relative decrease in the electrical conductivity will become smaller at higher electrolyte concentration in the liquid, which in turn, will improve the measuring accuracy.

4.2.2. Effect of particle blocking

The modified probe was used to measure the liquid-solid system to study whether the sieve will block fine particles. Experiments were carried out with different particle sizes (glass beads: 53, 83 and 117 μm), as shown in Fig. 17. The results show that the blocking of fine particles ($<83 \mu\text{m}$) is negligible in a liquid-solid flowing system. The experiments also showed that the modified probe was still applicable in a gas-liquid-solid

slurry system with small particles to measure the local solid holdup at high solid holdup up to 40%. However, when the particle size is relatively larger, the effect of blocking particles caused a decrease in the measured solid holdup and additional modification to Eq. (19) must be considered, which will be studied in our succeeding work.

The local solid holdup in the gas-liquid-solid slurry system can be measured on-line with the conductivity probe method, which is not available with the similar method in the literature. The measurement of the local solid holdup is important to provide more information for a better understanding of the

Table 2
The measured electrical conductivity with the original and modified probes in a gas-liquid system

	U_g (m/s)			
	0	0.0284	0.1420	0.2840
The original probe				
γ ($\mu\text{S/cm}$)	510.6	496.34	423.28	355.17
$\Delta\gamma$ ($\mu\text{S/cm}$)	–	14.26	87.32	155.43
$\Delta\epsilon_s$ (%)	–	4.13	23.63	39.63
The improved probe				
γ ($\mu\text{S/cm}$)	510.6	509.25	505.14	500.745
$\Delta\gamma$ ($\mu\text{S/cm}$)	–	1.35	5.46	9.855
$\Delta\epsilon_s$ (%)	–	0.39	1.59	2.86

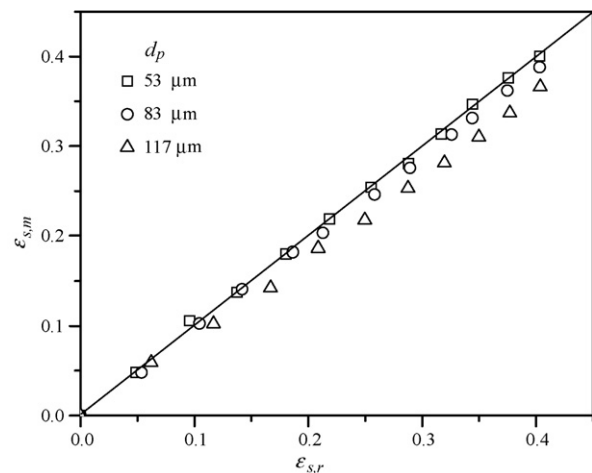


Fig. 17. Effect of blocking particles with the modified probes in a liquid-solid system.

hydrodynamics and mass-transfer behavior in a liquid–solid and gas–liquid–solid systems.

5. Conclusions

The electrical conductivity probe method for measuring the local solid holdup was studied both theoretically and experimentally. The following conclusions can be drawn:

- The relationship between the solid particles and the mixture electrical conductivity was deduced from one particle to multiple particles based on the electric field theory. This relationship can be used to measure the local solid holdup in a liquid–solid system.
- The developed model was validated by several cases of experiments. The electrolyte concentration in the liquid and the electrolyte adhered on particles have no effect on the measuring accuracy. However, the particle shape has significant effect on the measured results.
- The probe was modified by adding sieve properly for use in a gas–liquid–solid slurry systems and the measuring accuracy was only slightly influenced by the sieve added (60 mesh) when the size of particles less than 83 μm even at a high solid holdup up to 40%.

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Appendix A. General solution of the Laplace's equation in the spherical coordinate

The Laplace's equation is

$$\nabla^2\phi = 0 \quad (\text{A.1})$$

The form of Eq. (A.1) in the spherical coordinate is

$$\nabla^2\phi(r, \alpha, \theta) = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial\phi}{\partial r} \right) + \frac{1}{r^2 \sin\theta} \frac{\partial}{\partial\theta} \left(\sin\theta \frac{\partial\phi}{\partial\theta} \right) + \frac{1}{r^2 \sin^2\theta} \frac{\partial^2\phi}{\partial\alpha^2} = 0 \quad (\text{A.2})$$

Supposing $\phi(r, \alpha, \theta)$ is axisymmetrical, Eq. (A.2) is simplified as

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial\phi}{\partial r} \right) + \frac{1}{r^2 \sin\theta} \frac{\partial}{\partial\theta} \left(\sin\theta \frac{\partial\phi}{\partial\theta} \right) = 0 \quad (\text{A.3})$$

Using the method of variable separation and assuming Eq. (A.3) has the solution with the following form:

$$\phi(r, \theta) = R(r)\Theta(\theta) \quad (\text{A.4})$$

Substitution of Eq. (A.4) into (A.3) and multiplying the resulted equation by r^2 gives:

$$\frac{1}{R(r)} \frac{d}{dr} \left(r^2 \frac{dR(r)}{dr} \right) + \frac{1}{\Theta(\theta) \sin\theta} \frac{d}{d\theta} \left(\sin\theta \frac{d\Theta(\theta)}{d\theta} \right) = 0 \quad (\text{A.5})$$

The first item of the left-hand side of Eq. (A.5) has only the variable r , and the second item has only the variable θ . Since the equation stands for all r and θ , each item should be constant and their sum equals to zero, therefore:

$$\frac{1}{R(r)} \frac{d}{dr} \left(r^2 \frac{dR(r)}{dr} \right) = k^2 \quad (\text{A.6})$$

$$\frac{1}{\Theta(\theta) \sin\theta} \frac{d}{d\theta} \left(\sin\theta \frac{d\Theta(\theta)}{d\theta} \right) = -k^2 \quad (\text{A.7})$$

where k is the separating variable.

Eq. (A.6) can be arranged as

$$r^2 \frac{d^2 R(r)}{dr^2} + 2r \frac{dR(r)}{dr} - k^2 R(r) = 0 \quad (\text{A.8})$$

The general solution of Eq. (A.8) is

$$R(r) = A_n r^n + B_n r^{-(n+1)} \quad (\text{A.9})$$

where A_n and B_n are undetermined coefficients, which can be determined from boundary conditions.

Substitutions of Eq. (A.9) into (A.8) yields:

$$k^2 = n(n+1) \quad (\text{A.10})$$

where $n = 1, 2, 3, \dots$ is positive integer.

Substitution of Eq. (A.10) into (A.7) gives:

$$\frac{d}{d\theta} \left(\sin\theta \frac{d\Theta(\theta)}{d\theta} \right) + n(n+1) \sin\theta \Theta(\theta) = 0 \quad (\text{A.11})$$

Eq. (A.11) is called Legendre equation, and its solution is Legendre function $P_n(\cos\theta)$:

$$\Theta(\theta) = P_n(\cos\theta) \quad (\text{A.12})$$

with $P_0(\cos\theta) = 1$, $P_1(\cos\theta) = \cos\theta$, $P_2(\cos\theta) = (3\cos^2\theta - 1)/2$, etc.

Substitution of Eqs. (A.9) and (A.12) into Eq. (A.4), the general solution of the Laplace's equation in the spherical coordinates is obtained:

$$\phi_n(r, \theta) = [A_n r^n + B_n r^{-(n+1)}] P_n(\cos\theta) \quad (\text{A.13})$$

The entire solution can be expressed by the sum of n Legendre polynomial as

$$\phi(r, \theta) = \sum_{n=0}^{\infty} [A_n r^n + B_n r^{-(n+1)}] P_n(\cos\theta) \quad (\text{A.14})$$

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