1 Introduction

Nowadays, the study of the properties of particulate systems is becoming increasingly important and necessary in numerous processes involving the production, handling and processing of particles, so as to improve the efficiency of these systems and to permit their control. Particle size and concentration affect the properties of a particulate system in many important ways, such as the taste of food and the colour of pigments. In [1], many methods are introduced to measure particle size. The choice of methods depends on the particle properties and the situations of the real applications. For example, optical methods, such as light scattering, absorption, transmission or extinction, have the distinct advantage that the measurements can be made without disturbing suspensions and sometimes the tests can be carried out in a form which lends itself to automatic recording and remote-control techniques. This makes possible nondestructive and nonintrusive process control and product quality measurement.

In traditional extinction measurements, when a light beam falls upon a particle suspension, the attenuation of the light intensity can be described with Bouguer-Lambert-Beer‘s law (BLBL) [2-3]. Over the recent two decades, dynamic extinction measurements have been developed further by many researchers [4-18]. Among them, Gregory [4] measured the mean particle size and concentration from the turbidity fluctuations. Wessely et al. [5-6] determined the particle size distribution (PSD) by varying the beam size. Based on the statistical characteristics of the transmission fluctuations in flowing particle suspensions, Riebel’s research group [7-18] developed a new method called transmission fluctuation spectrometry (TFS) for particle size analysis by means of a non-linear operation on the transmission fluctuations T, e.g., logarithm of transmission (lnT), expectancy of the transmission square (ETS). The TFS measurement by means of the ETS was realized with a combined temporal and spatial averaging of the transmission signals through the flowing particle suspension, provided that the flow velocity is known. Signal averaging was executed in the frequency domain with an array of 1st order low pass filters. In order to reduce or avoid the influence of the flow conditions, another approach based on TFS but independent of the flow conditions is worth putting forward and further investigating. Apart from this, the resolution of measurements is also worthy of improvement.

The main aim of this thesis is to introduce a new approach called transmission fluctuation correlation spectrometry (TFCS), which offers a steep transition function and hence a high resolution for particle size analysis. Another aim is to realize measurements on flowing particle suspensions, independent of the flow conditions. Furthermore, local turbulent multiphase flow structures are investigated by means of the TFCS technique in combination with the Laser Doppler Velocimetry (LDV) technique. Some of the results have already been published partly in journals and conference proceedings (see publication list). The objective is to summarize the results, present some new results and discuss some aspects in more detail. The work presented here is constructed as follows.

In chapter 2, a brief review of the previous state of the TFS technique is given. The analytical expressions for the ETS with different combinations of temporal and/or spatial averaging are applied to particle size analysis. Spatial averaging depends on the beam profile, such as an infinitesimally thin beam, a circular uniform beam and a circular Gaussian beam. Temporal averaging can be carried out in the time domain with variable gliding time intervals or in the frequency domain with the 1st order low pass filters at various cutoff frequencies.

Chapter 3 presents the theory on the TFCS. In view of the concentration effects of hard-core spherical particles in the suspension, a direct correlation function (DCF) of two-dimensional hard sphere fluids is proposed theoretically. Based on the layer model, theoretical description of the TFCS in terms of the expectancy of the transmission product (ETP) is provided under the assumptions of geometrical ray propagation and opaque particles in the suspension. Spatial correlation (TFS-SC) is realized by changing the distance between dual parallel beams and autocorrelation (TFS-AC) can be realized by varying the correlation time with a single beam. Then the theory is extended to the case of crossed beams as well as to the case of partially transparent particles with penetrating radiation.

In chapter 4, numerical simulations on the two-dimensional (2D) monolayer and three-dimensional (3D) suspension are executed, also assuming geometric ray propagation and hard-core particles. Simulations offer the possibility of studying the effects, which arise from particle concentration or monolayer density and from the structure due to steric particle-particle interactions, on the extinction and transmission fluctuation correlation spectrum.

Detailed experimental techniques are described in chapter 5, where the experimental systems of TFCS and LDV are given and applied to measure the flowing particle suspensions and local flow structures.

In chapter 6, the relationship between the particle size, concentration and transmission fluctuations is investigated experimentally by using a narrow laser beam. The deviation from BLBL in concentrated particle suspensions is corrected with three various approaches including Riebel's steric particle-particle interaction model, Vereshchagin's shielding effect approximation and a polynomial fit of particle concentration.

Two iterative inversion algorithms, i.e., modified Chahine inversion [19] and relaxation method [20-21], are studied in detail in chapter 7. They are applied to retrieve the information on particle size distribution and particle concentration in the TFCS measurements. The convergence, accuracy and stability of the inversion algorithms are tested with numerically simulated data containing different levels of noise and realistic experimental data.

Chapter 8 presents the TFCS measurements on the flowing particle suspensions. A narrow focused Gaussian beam is employed in the TFS-AC technique. The effect of the monolayer density on the spatial structure of the particle suspensions is investigated experimentally, compared with both the theoretical predictions in chapter 3 and the numerical simulations in chapter 4. Furthermore, the flowing particle suspensions are measured by TFS-SC, whereby the experimental transition spectrum is obtained by changing the distance of bibeam instead of by varying the autocorrelation time.

In chapter 9, the characteristic feature of vortical turbulent multiphase flow structures behind a bluff body is investigated by means of the TFCS technique in combination with the LDV technique. Vortex shedding frequency, seed particle size, concentration and local vortex flow structure dimension are measured. The 3D experiment is compared with a 2D numerical simulation. As a comparative study, the pulsating flow is also studied in this chapter.

Some general conclusions are drawn in the last chapter, with some outlooks on the present transmission fluctuation spectrometric study discussed.

2 Brief Review of Transmission Fluctuation Spectrometry

2.1 Introduction to Transmission Fluctuations

When a beam of light is incident upon a particle suspension, a portion of the light interacts with the particles inside the beam. These interactions involve diffraction, scattering and absorption. The transmitted light intensity attenuates, depending on the particle properties. The transmission T of the beam is defined as

$$T = \frac{I}{I_0}, \tag{2.1}$$

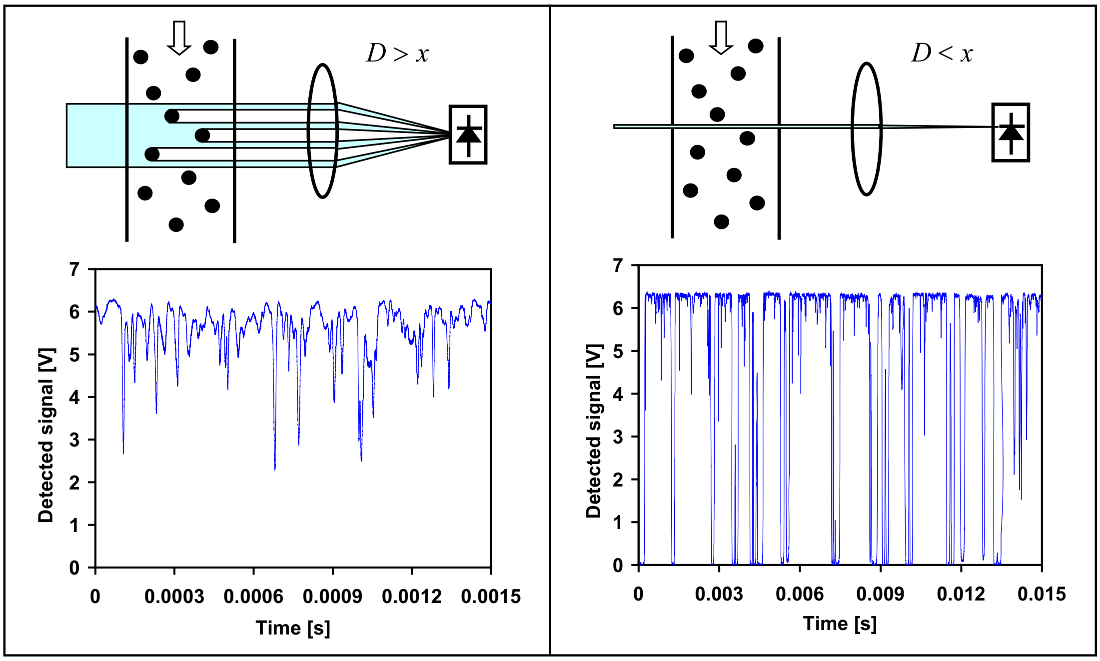
where I, I_0 are the intensities of the transmitted light in the presence and absence of particles, respectively. The extinction E is generally described by Bouguer-Lambert-Beer's Law (BLBL) [2-3]. For a suspension of monodisperse spherical particles, BLBL is written as follows.

$$E_{BLBL} = -\ln T = \left(\alpha_p - \alpha_f\right) C_V \Delta Z = K_{ext} C_{PA} \Delta Z \tag{2.2}$$

where α_p and α_f are absorption coefficients of the particle material and the suspension fluid, respectively. ΔZ is the optical path length of the suspension, K_{ext} the extinction efficiency of a single particle, C_{PA} and C_V are particle projected area concentration and volume concentration, respectively.

It is worth mentioning that the BLBL is derived by integration from an intensity balance for an infinitesimally thin layer of suspension. This derivation is based on the following inherent assumptions [22]. Firstly, radiation energy is assumed to propagate unidirectionally only, that is, the effect of multiple scattering is negligible. Secondly, the particles are assumed to scatter radiation independently of neighbouring particles, that is, K_{ext} is assumed to be independent of particle concentration and effect of dependent scattering is also negligible. Furthermore, in order to satisfy the integrability condition, the intensity attenuation in each thin suspension layer must be infinitesimally small or at least much smaller than the intensity itself. At the same time, the thickness of the layer in question must be superior to the particle diameter. Hence only in the limit of low particle concentrations does the BLBL hold true. At high particle concentrations, the signals will be modified by steric particle-particle interactions.

Based on a quasi-continuum approach, the BLBL does not account for the discrete nature of particles or their spatial extension and arrangement. However, in practical extinction measurements on the flowing suspensions, the discrete nature of particles leads to time-dependent transmission fluctuations $T(t)$, especially when the beam diameter D is not large enough or even smaller than the particle diameter x. Figure 2.1 illustrates this effect.



(a) $D > x$. (b) $D < x$.

Fig.2.1 Transmission fluctuations produced by flowing suspensions. (a) Wide beam $D > x$; (b) Narrow beam $D < x$.

Figure 2.1a portrays the "near-BLBL case", where the beam is sufficiently large to cover several particles which altogether make up the measured transmission. Due to the almost negligible contribution of a single particle, the detected transmission shows a weak fluctuation. An averaging procedure or a low-pass circuit is used in general to eliminate the fluctuations and extract the information on particle concentration with Eq.(2.2). When the beam diameter is much smaller than the particle diameter shown in Fig.2.1b, a single particle can block the light and hence the transmission signal fluctuates violently between 0 and 1 with the assumptions of geometric ray propagation and perfectly absorbent particles.

The transmission fluctuation signals detected in a long period of time t_s can be treated, resulting in the average transmission or the expectancy of transmission (ET), standard deviation σ_T and the expectancy of the transmission square (ETS).

$$ET = \lim_{t_s \to \infty} \frac{1}{t_s} \int_{t_0}^{t_0+t_s} T(t) dt \tag{2.3}$$

$$\sigma_T = \sqrt{\lim_{t_s \to \infty} \frac{1}{t_s} \int_{t_0}^{t_0+t_s} [T(t) - ET]^2 dt} \tag{2.4}$$

$$ETS = \lim_{t_s \to \infty} \frac{1}{t_s} \int_{t_0}^{t_0+t_s} [T(t)]^2 \, dt = (ET)^2 + \sigma_T^2 \tag{2.5}$$

The ET and σ_T can be used to measure particle concentration and mean particle size [4]. By varying the beam diameter, the particle size distribution can be measured [5-6]. The ETS with the spatial and/or temporal averaging in the time domain or frequency domain can be applied to particle size analysis [8, 10-17]. A brief review of the previous TFS realized by the ETS is given in the following sections.

2.2 Basic Considerations

Theoretical descriptions of the ETS are based on the assumptions of geometrical ray propagation and perfectly absorbent particles. The interaction of the radiation with the particles is modeled as the blockage of straight rays. As illustrated in Fig.2.2a, a three-dimensional monodisperse suspension is modeled as a pile of N_{ML} statistically independent monolayers [11, 13, 22]. The thickness ΔZ_{ML} of a monolayer is taken to be at least one particle diameter x:

$$\Delta Z_{ML} = \frac{P}{1.5} x , \tag{2.6}$$

where P is a structure parameter varying between 1.5 and 2.9, depending on the flow regime. The subscript "ML" denotes monolayer.

The total number of monolayers is obtained as

$$N_{ML} = \frac{\Delta Z}{\Delta Z_{ML}} = \frac{1.5}{P} \cdot \frac{\Delta Z}{x} . \tag{2.7}$$

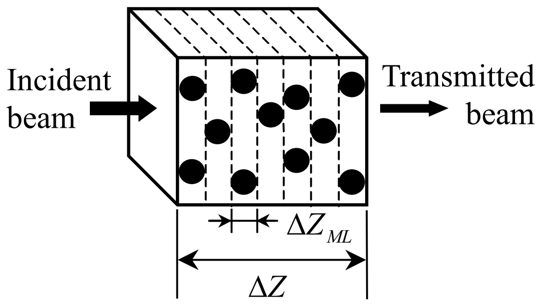
The transmission T through the whole suspension is the product of the transmissions through the individual layers, taking the form [22, 24]

$$T = \prod_{i=1}^{N_{ML}} T_{ML,i} = (1 - \beta K_{ext})^{N_{ML}} , \quad \beta K_{ext} << 1 \tag{2.8}$$

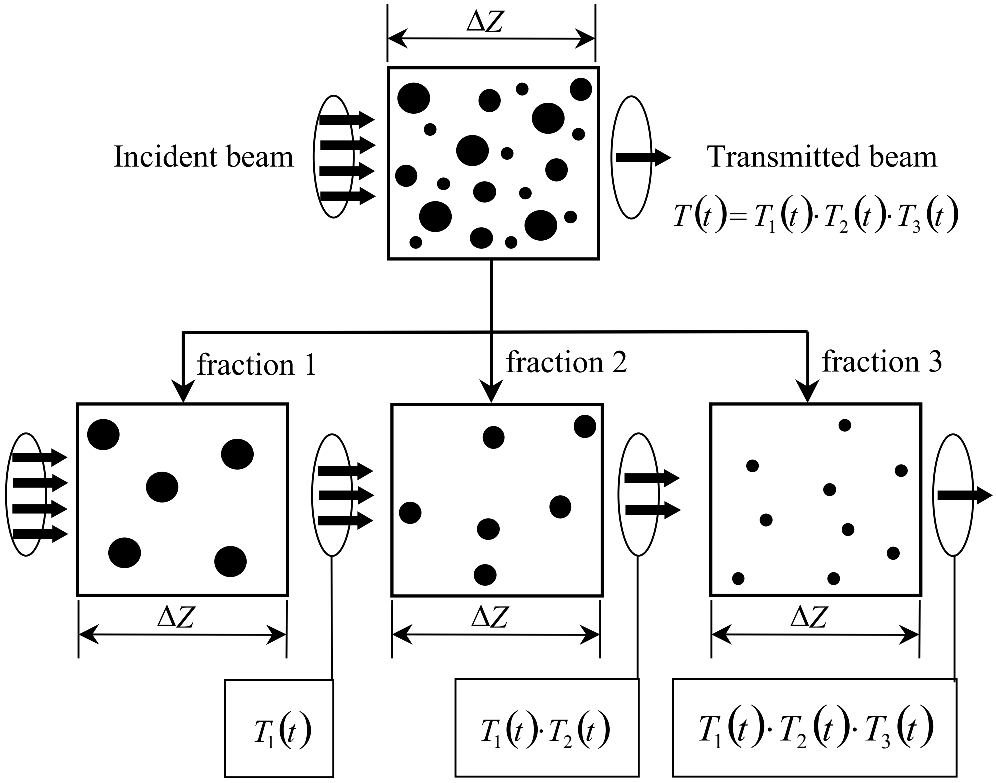
where $\beta = PC_V$ is the monolayer density, defined as the ratio of the projected area covered by particles in the monolayer to the total area of the monolayer.

In a similar manner, the ETS of a three-dimensional suspension can be given by [10]

$$ETS = \prod_{i=1}^{N_{ML}} e\{T_{ML,i}^2\}. \tag{2.9}$$



(a) Multilayer transmission model.



(b) Polydisperse suspension.

Fig.2.2 Layer model for the suspension [11, 13, 22].

With Eqs.(2.6-2.8), the extinction based on the layer model is expressed as

$$E = -\frac{1.5}{P} \cdot \frac{\Delta Z}{x} \cdot \ln\left(1 - P C_V K_{ext}\right). \tag{2.10}$$

The deviation of extinction from the BLBL is corrected by a factor $F\left(K_{ext}, C_V\right)$ due to steric particle-particle interactions with increasing particle concentration [9, 22-23]

$$E = F\left(K_{ext}, C_V\right) \cdot E_{BLBL}\,, \tag{2.11}$$

with

$$F(K_{ext}, C_V) = \frac{\ln(1 - PC_V K_{ext})}{-PC_V K_{ext}}. \quad (2.12)$$

As a result, the statistical behavior of the whole suspension may be modeled on the basis of a single monolayer.

In addition, a dilute polydisperse suspension may also be modeled as a pile of monodisperse suspensions, on condition that particle positions are independent of each other, i.e., steric particle-particle interactions are negligible. As shown in Fig.2.2b, the incident light is modulated by a series of cascaded volumes, each containing a monodisperse suspension. The transmission after the first volume is $T_1(t)$ and the transmission after the second volume is the transmission of the first one modulated by the fluctuation of the second system, which is $T_1(t) \cdot T_2(t)$. The rest may be deduced by analogy. Thus the total transmission is given by the product of all single transmissions. This is also valid for the ETS [10].

2.3 ETS with Temporal Averaging

At an early stage, transmission fluctuation spectrometry was studied by Kräuter et al [8, 10] by means of an infinitesimally thin beam through a flowing suspension. Particle extinction efficiency K_{ext} is 1.0 assuming geometric ray propagation. Due to the blockage of light by the discrete opaque particles, the transmission signal $T(t)$ statistically jumps in a binary manner between $T = 0$ and $T = 1$, resulting in

$$ET = ETS = 1 - \beta. \quad (2.13)$$

A gliding averaging with a variable time constant τ leads to the averaged signal $T_\tau(t)$

$$T_\tau(t) = \frac{1}{\tau} \int_{t_0}^{t_0+\tau} T(t) dt. \quad (2.14)$$

Two special limit cases are taken into consideration as follows.

$$\lim_{\tau \to 0} T_\tau(t) = T(t) \quad (2.15)$$

$$\lim_{\tau \to \infty} T_\tau(t) = ET \quad (2.16)$$

With increasing the averaging time constant τ, a transition of the ETS occurs between the limits:

$$\lim_{\tau \to 0} e\{T_\tau^2(t)\} = ETS = 1 - \beta, \quad (2.17)$$

$$\lim_{\tau \to \infty} e\{T_\tau^2(t)\} = (ET)^2 = (1 - \beta)^2. \quad (2.18)$$

A theoretical description of the ETS through a monolayer with temporal averaging is given by