Separation of gas-borne nanoparticles in bubble columns

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A B S T R A C T

The separation efficiency of bubble columns was studied for gas-borne nanoparticles with different properties ranging from sodium chloride (NaCl) spheres to carbon agglomerates. In addition, the influence of the concentration of a polymeric stabiliser (polyvinylpyrrolidone (PVP) K25) on the separation efficiency was investigated. For pure water, it was found that the experimentally determined separation efficiencies were well recovered by employing the Fuchs model based on the measured relative bubble rise velocities. In particular, it was shown that the particles exhibit a sticking probability of one at the gas–liquid interface. With increasing stabiliser concentration, the gas–liquid interfaces become increasingly rigid, finally leading to the complete cease of internal circulation inside the bubble. Then, the Fuchs approach is no longer valid and a model based on pure Brownian diffusion applies. This so called Friedlander model was capable of quantitatively predicting the separation efficiencies. Beside the larger bubbles, very small bubbles were formed when using PVP solutions. The extremely high separation efficiencies of these microbubbles may indicate a new way for high performance nanoparticle deposition in bubble columns.

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1. Introduction

The transfer of particles from the gas phase into a liquid in bubble columns has been employed intensely for gas cleaning purposes. Two decades ago, special interest on this gas cleaning topic emerged from the retention of radioactive aerosol in wet scrubbers as reflected in literature (Jonas & Schütz, 1988; Pich & Schütz, 1991; Ghiaasiaan & Yao, 1997). Also the removal of fly ash particles by bubble columns was investigated (Meikap & Biswas, 2004). In general, the exponential increase of the separation efficiency with column height was found, similar to the increase of separation efficiency with thickness of fibrous filters. However, since dealing with micron aerosol particles, these studies considered impaction as dominant transport mechanism of the particles from the gas to the liquid boundary. Moreover, the investigations were performed rather on column systems close to industrial applications showing highly turbulent bubble flow prone to broad distributions of residence times and velocities. In addition, in order to optimise separation efficiencies high bubble concentrations were intended possibly resulting in bubble coalescence. On the separation of aerosol nanoparticles in bubble columns, much less is known. Due to the likely impact of these particles on public health (Oberdörster, 2000), efficient gas cleaning strategies for nanoparticles are searched. In contrast to filters, which show increasing pressure drops during operation eventually leading to clogging (Thomas et al., 2001), bubble columns can be operated for extended periods of time at constant pressure drop. But the reported separation efficiencies vary to a large extent and the particle deposition lacks a quantitative description. Based on separation experiments of organic particles in impingers with a fritted nozzle tip, Miljevic et al. (2009) found a weak size dependency of the separation efficiency in water and in heptane

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in some cases, in other cases they did not observe any systematic size dependency. Charvet et al. (2011) studied the separation of carbon nanoparticles in a water bubble column with 12 inlet nozzles. They found a most penetrating particle size in the range between 10 and 30 nm. However, unlike Miljevic et al. (2009), they did not take into account residual particles originating from the bursting of bubbles at the liquid surface of the bubble column. In addition, their model was only capable of fitting the experimental data over a very limited size range. None of these studies considered the fact that the particle deposition efficiency depends on both, absolute and relative bubble velocity, respectively. While the absolute bubble rise velocity controls the residence time, the relative velocity between bubble and surrounding liquid determines the internal circulation. Especially, the internal circulation (e.g. in form of a Hill vortex) influences the contribution of convective and diffusive nanoparticle transport from the gas phase to the liquid boundary.

Therefore, the first goal of this paper is to give a closed and validated model for the nanoparticle separation in water bubble columns. In particular, it will be shown that particles stick to the gas–liquid boundary at first contact independent of particle structure and hydrophobicity. By using the correct relative velocities, this model gives a quantitative description of the separation efficiency based on the particle mobility equivalent diameter. Based on experimentally determined mass balances, separation efficiencies for spherical sodium chloride particles and carbon agglomerates in pure water are compared with a simple model introduced by Fuchs (1964), which accounts for the interference of various deposition mechanisms working in parallel such as Brownian diffusion, convection, sedimentation and inertial motion, respectively. The second goal of the paper is to include the influence of surfactants employed to obtain stable nanoparticle suspensions (Verma et al., 2009). This was the original starting point of this work. In an attempt to conserve the special size-dependent properties of nanoparticles, like substantially enhanced bioavailability of pharmaceutical and nutrition products compared to larger particles (Hu et al., 2004), Rapid Expansion of Supercritical Solutions into Aqueous Solutions (RESSAS) may be employed (Türk et al., 2002a, 2002b). In the RESSAS process, a supercritical solution is expanded into a liquid which contains surfactants in order to stabilise the particles in an early stage of growth leading to the desired stable nanosuspension (Myers, 1991; Patrick et al., 1997). In principle, the transfer of nanoparticles into a liquid consists of their gas phase transfer to the bubble boundary (convective flux), the adsorption of surfactant molecules to the particle surface and the transport across the interface into the bulk liquid (diffusive flux) as depicted later in Fig. 2. However, the scope of this paper will be restricted to the gas phase transport of the particles and the sticking to the liquid boundary. Depending on particle material and surfactant type, high separation and stabilisation efficiencies have been observed in RESSAS experiments (Türk et al., 2002a, 2002b). However, to the best of our knowledge, the influence of surfactants added to the liquid in the bubble column has not been studied systematically with regard to the separation efficiency. This could in principle be done on a RESSAS apparatus. However, due to involved hydrodynamics including highly turbulent multiphase flow and bubble coalescence, this set-up will not allow to clarify the basic mechanisms of nanoparticles transfer from the gas phase into the liquid.

Therefore, the approach in the present work is to study the separation efficiency of nanoparticles in bubble columns systematically as a function of different parameters in a simplified model system. The influence of particle material, morphology and size as well as the impact of surfactant concentration on the separation efficiency are examined and discussed here. In the model bubble column, a well-defined aerosol was injected through a nozzle at low flow rates, so that single, mostly spherical bubbles with a narrow size distribution were rising in a much less turbulent flow than in the RESSAS column, thus, allowing to follow the path of individual bubbles. The effect of reduced internal circulation in the bubbles by the addition of the surfactant to the liquid is measured for different concentrations of polyvinylpyrrolidone (PVP) K25. At higher PVP concentrations, where the gas inside the bubble is stagnant (no internal circulation), an approach to calculate the separation efficiency proposed by Kronig & Brink (1951) and discussed by Friedlander (2000) is employed.

2. Theoretical aspects and background

The internal circulation in the rising bubble has a major influence on the particle separation. For diffusional deposition, the internal flow pattern defines the vicinity to which the particles approach the bubble boundary by convection, while for inertial deposition the circulation intensity determines the centrifugal forces acting on the particles (Fig. 1). Therefore, in the theoretical section, the Fuchs model for aerosol particle deposition in rising gas bubbles will be outlined first, then the influence of stabilising detergents on the internal circulation and on the resulting deposition efficiency will be discussed, and, finally, the effect of bubble concentration on the relative rising velocity will be reviewed.

2.1. Fuchs model for the aerosol deposition in rising gas bubbles

Fuchs (1964) introduced a deposition model in order to describe the absorption of aerosol particles during the bubble rise in a liquid. Thereby, Fuchs neglected the influence of surface-active impurities in the liquid, which will be taken into account later in this work. Fuchs assumed that the internal circulation inside the rising gas bubble represents a Hill's vortex without retardation so that the tangential surface velocity at bubble equator u_S is related to the bubble rising velocity u_B by $u_S = \frac{3}{2} u_B$. The internal circulation is shown in Fig. 1 and is induced by the fluid flow friction on the gas–liquid interface. The main region for the diffusional deposition of particles is the equatorial region, while for inertial deposition the particles experience the largest centrifugal forces in the strong curvatures towards the poles.