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Enhancement of gas-to-liquid oxygen transfer in the presence of fine solid particles for air-exposed multiphase system

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ABSTRACT

To investigate “particle effect” promoting gas-liquid mass transfer for a better process control, nine species of fine particle ($d_{32} = 1\text{--}15\ \mu\text{m}$), divided into Group 1 (SSA > 65.4 m²/g, termed PAC, SiO₂, Al₂O₃ and MnO₂) and Group 2 (SSA < 9.8 m²/g, termed Biochar, Graphite, Mg(OH)₂, CaCO₃ and BaSO₄), were comparatively introduced for oxygenation tests in an air-exposed stirred system. Group 1 particles with interfacial-cleaning effect facilitated oxygen transfer better than their counterparts of Group 2. For all the particles employed, however, the “particle effect” interestingly presented under control of solid specific gravity, since enhancement factor (E_f) globally performed to grew up either when particulate density decreased or liquid density increased (Pb(NO₃)₂ as solute). Moreover, the strength of “particle effect” depends largely on interfacial turbulence, because proper agitation allowed optimal acceleration of oxygen transfer (700 rpm for this work), while either more sluggish or turbulent flow regime would hinder or even eliminate “particle effect”. Consequently, interfacial liquid-entrainment of particle entities was assumed to have played an initiative role for induction of “particle effect”, and this hydrodynamic process was elucidated to be subject to particulate specific gravity as well as interfacial turbulence.

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1. Introduction

Multiphase flow often takes place in chemical and biochemical industries consuming specific gaseous species, where suspended solid media are usually employed as heterogeneous reactant, functionalized catalyst or active substrates (Lindner et al., 1988). Mechanical agitation has been frequently utilized for mixing and mass transfer, because it is simple and flexible in process control. Prior to intense bulk reaction, however, a sufficient gas-to-liquid mass transfer is required to meet its fast consumption which is commonly the overall

rate-limiting step (Joly-Vuillemin et al., 1996). For example in environmental practices, the oxygen transfer dominates the aerobic treatment of wastewater in a stirred-tank bioreactor (Zokaei-Kadijani et al., 2013). In fact, fine solid particles, usually present as catalyst or facilitator for bulk reaction, may simultaneously play a role to improve gas-liquid mass transfer (Dagaonkar et al., 2001) that can be labeled as “particle effect”.

The interesting nature of solid particles inducing “particle effect” has drawn a great deal of concern by researchers. It was documented that the addition of fine particles in micron-sized (μm) level (similar to the order of liquid-film thickness)

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could give rise to an enhanced physical gas-absorption rate, whereas particles with larger size almost had no effect (Zhang et al., 2008). The solid loading is one of influencing factors as Smith and Skidmore (1990) have found that volumetric gas transfer coefficient increased with solid fraction of coal up to 2 wt% and then began to decrease, while Hwang and Lu (1997) reported a net reduction of gas transfer rate with an increase in amount of polystyrene particle.

Mechanism study aiming to interpret the particle-induced enhancement effect on gas transfer has been conducted since the original work published by Alper et al. (1980). Some researches attributed the particle effect to an adsorptive transport notion that was called “shuttle and grazing effect”, for which particles were assumed to adsorb an excessive amount of gas at the interface or within liquid film and then release the adsorbed molecules into the liquid bulk (Alper et al., 1980; Tinge and Drinkenburg, 1992; Demmink et al., 1998; Ruthiya et al., 2003). More recently, the effect of solid particles has been suggested due to the adsorptive elimination of trace interfacial “contaminants” such as dust and surfactants that hinder gas-liquid mass transfer (Svitova et al., 2001; Levitz, 2002; González-García et al., 2004; Kaya and Schumpe, 2005; Kordač and Linek, 2006; Rosu and Schumpe, 2007). Kordač and Linek (2006) indicated that the interfacial boundary layer is “rigid” without particles that locally inhibits the mobility of liquid along the interface, while in the presence of particles it is mobile and the surface renewal proceeds (the explanation is very close to that suggested by Kaya and Schumpe (2005)). Yet Garcia-Ochoa et al. (1997) in their research noticed that the presence of pyrite particles (not porous) resulted in higher volumetric mass transfer coefficient compared to fine glass beads, which seems not to be in accordance with the “surfactant grazing” effect. Vinke et al. (1993) proposed a model with assumption of adhering particles relating the enhancement of incipient gas absorption which was verified by flotation experiments and gas-absorption measurements. Opposite conclusion against “particle effect” has also been reported as Derksen et al. (2000) observed a diminution of gas-liquid mass transfer coefficient once hydrophobic silica particles ($d_{50}=13\ \mu\text{m}$) were added in a stirred tank reactor which was ascribed to interfacial block effect.

With respect to “particle effect” in a stirred system actually, explanations made solely by physicochemical properties of particles may not be comprehensive enough, since the motion of solid suspensions influenced by liquid turbulence should be considered as well, for which a transient impact of particles accumulating at gas-liquid interfacial region may be evoked affecting gas transfer. Roizard et al. (1999) observed that under flow conditions activated carbon ($\leq 20\ \mu\text{m}$) investigated could not adhere steadily at the gas-liquid interface, and they made a recommendation that enhancement of mass transfer cannot be simply explained by stagnant adhesion of particles. That is to say, the hydrodynamic behavior of solid individuals should be taken into consideration on interpreting the “particle effect”. Similarly, Kluytmans et al. (2003) postulated that the solid particles should have played a back-mixing role on boundary layer thus improved gas transfer. They suggested that the increment of stirring intensity was not favorable for “particle effect” (using activated carbon), since in response the ratio of mass transfer coefficient with and without particles would begin to decrease. All the scientific efforts indicated that a more likely explanation for “particle effect” should be a collection of influencing factors including both the relevant particulate properties and hydrodynamics, and on this regard,

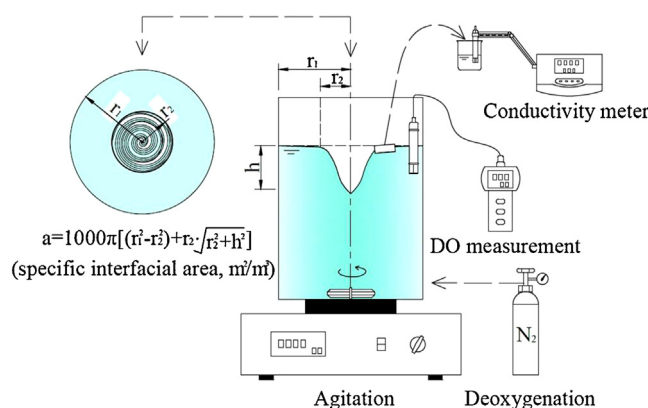


Fig. 1 – Schematic diagram of experimental setup.

the controlling parameters accounting for “particle effect” needs to be further specified and investigated in multiphase flow.

In this paper, comparative investigation on kinetics of oxygen transfer was conducted using nine species of fine particle (deliberately selected), for which main purposes were (1) to extract the controlling factors for occurrence of “particle effect”, and (2) to make further discussion on possible mechanism interpreting the functionalization of fine particles as promoter for gas-liquid mass transfer.

2. Materials and methods

2.1. Materials

Powdered activated carbon (PAC), calcium carbonate (CaCO_3 , analytical reagent) and manganese black (MnO_2 , analytical reagent) were purchased from Kermel Reagent Co. Ltd. (Tianjin, China). Fine graphite powder (Graphite, analytical reagent) and aluminum oxide (Al_2O_3 , analytical reagent) were supplied by Sinopharm Chemical Reagent Co. Ltd. (Shanghai, China). Magnesium hydroxide ($\text{Mg}(\text{OH})_2$, analytical reagent) was obtained from Fuchen Chemical Reagent Factory (Tianjin, China). Barium sulfate (BaSO_4 , analytical reagent) was provided by Silian Chemical Plant Co. Ltd. (Shanghai, China). Silicon dioxide (SiO_2 , analytical reagent) was bought from Ruijinte Chemical Reagent Co. Ltd. (Tianjin, China). Biochar particles were produced in laboratory by means of hydrothermal carbonization according to Xiao et al. (2012), for which poplar sawdust was utilized as raw material. Plumbous nitrate ($\text{Pb}(\text{NO}_3)_2$, analytical reagent) was purchased from Kaixin Chemical Reagent Co. Ltd. (Tianjin, China). Purified water was employed as liquid phase that was prepared by local tap water undergoing filtration and ion exchange (resin column).

2.2. Oxygen transfer experiments

Oxygen transfer tests were performed in a cylindrical glass reactor (unbaffled) with internal diameter of 120 mm containing 1 L liquid as illustrated by Fig. 1. Digital tachometer magnetic stirrer (Jiangsu, China) was utilized to control the stirring speed in the range from 100 to 1000 rpm. Prior to each oxygen transfer test, a preset amount of solid sample used was milled using agate mortar, ultrasonically dispersed in water for 5 min, and then kept immersed for 1 h to ensure a complete wetting. The de-oxygenation as a preparation step for oxygen transfer was accomplished by bubbling of pure nitrogen gas. It should be addressed that for all the tests in the presence of

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