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Evaluation of a novel gas-liquid contactor/reactor system for natural gas applications





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ABSTRACT

In this study, a novel system for contacting gases and liquids, which may be suitable for many applications involving gas-liquid contact or gas-liquid reactions, has been developed and characterized. The system consists of a vertical vessel with gas and liquid ports and inert particles. The gas is injected through a single orifice at the bottom of the vessel and leaves at the top, while the liquid feed is introduced at the top, and the effluent leaves at the bottom of the vessel through a vertical tube inserted from the top all the way to the bottom. The system involves the use of inert particles that create circular motion within the vessel, enhancing mixing and providing high gas—liquid interfacial area for effective mass transfer. The contactor system was evaluated for the capture of CO_2 through reactions with ammonium hydroxide and saline wastewater, namely desalination reject brine. The effect of inert particles surface area on both CO_2 capture and ions removal was evaluated. The liquid residence time distribution (RTD) was also evaluated for experiments with and without the mixing particles, using sodium hydroxide as a tracer. In addition, the system dynamic behavior was assessed through step changes in the gas flow rate, liquid flow rate and temperature. The reactor system showed high reaction efficiency, excellent mixing and very stable steady state.

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1. Introduction

Gas-liquid contactors are widely used in many industries that involve such processes as chemical, petrochemical, biochemical, and metallurgical. The selection as well as the design, effectiveness and the overall performance of these contactors or reactors often depend on the transport mechanisms as well as contactor hydrodynamics and reaction kinetics. These units are commonly encountered as aerators or gas-liquid reactors, where the gas first dissolves in the liquid and then reacts with the liquid or any materials dissolved in the liquid. Reactions in these types of reactors are often divided into two categories: slow or fast reactions (Charpentier, 1981). Good mass transfer and high liquid hold-up are often needed to keep the gas bulk concentration near saturation for slow reactions, while high gas hold-up and very small gas bubbles are required in fast reactions, since the concentration of the gas in

* Corresponding author. E-mail address: muftah@qu.edu.qa (M.H. El-Naas). the bulk liquid is negligible and the rate of gas absorption is "controlled by gas—liquid interfacial area" (Charpentier, 1981). Higher mass transfer rates and correspondingly faster reaction rates can be achieved through enhancing the contact area between the liquid and the gas.

One major drawback of some high performance gas—liquid contactors that require good mixing is the need for high mechanical energy. However, such mechanical energy may be utilized more efficiently in some types of gas—liquid contactors than others. Gas—liquid contactors with diverse designs may exhibit significantly different mass transfer performance under the same operating conditions (Gaddis, 1999). Bubble column reactors, spouted bed reactors, and agitated reactors are known to be more suitable for processes involving slow reactions, such as liquid-phase oxidation, hydrogenation, chlorination and fermentations (Deckwer and Schumpe, 1993). Other types of gas-liquid reactors such as packed columns and venturi reactors are known to be more effective for processes with fast reactions, because of their high gas—liquid interfacial contact area (Deckwer and Schumpe, 1993); however, under certain conditions, bubble column reactors and packed column reactors can be suitable for processes with highly exothermic fast reactions, "which are widely used in the chemical, biochemical, petrochemical and metallurgical applications" (Deckwer and Schumpe, 1993). Bubble column reactors have several advantages, including low maintenance and operating cost, low capital cost, and excellent heat transfer and temperature control. They also tend to have high gas—liquid interfacial area as well as high volumetric mass transfer coefficient. These reactors, however, suffer from some drawbacks such as back-mixing and bubble—bubble interactions in the turbulent flow regimes. In addition, the lack of specific data on the reactor hydrodynamics and the characteristics of mass transfer under real industrial conditions makes it rather difficult to scale-up (Lemoine et al., 2008).

Gas-liquid contactors are often classified into surface and volume contactors. They may also be sorted in terms of mass transfer rate. In general, contactors with low mechanical energy consumption have low mass transfer rates and, consequently, low performance. More mechanical energy consumption, which is usually associated with more mixing inside the contactor, improves the mass transfer rate. Surface contactors are typically used for the bio-treatment of several types of wastewater and usually have the form of pools with moderately low depth. They often involve the use of impellers or liquid jets to enhance mixing and provide the needed interfacial area (Gaddis, 1999). On the other hand, in volume gas-liquid contactors, the contact area between the gas and liquid phases is often formed within the bulk of the liquid. Gas dispersion in the liquid, in the form of bubbles with spherical or irregular shapes, is usually achieved through the use of mixing tools such as "spargers, liquid jets, two-mixture nozzles or hollow rotating mixers" (Gaddis, 1999). Examples of common gas-liquid contactors/reactors include bubble column reactors, stirred vessel reactors, jet loop reactor, reciprocating jet reactor, and impingingstream reactor.

A bubble column reactor is a vessel with many shapes, where gas bubbles are created at the bottom of the vessel through a sparger. Due to their mechanical simplicity, relatively low capital cost, and good heat and mass transfer performance, bubble columns are widely used in many applications in bio-processing industry such as aerobic fermentation (McClure et al., 2015). The mass transfer coefficient in bubble columns is known to depend on such factors as the physical properties of the gases and liquids used, the gas flow rate (Gaddis, 1999), sparger design (Camarasa et al., 1999), reactor height to diameter ratio (H/D) (Anderson and Quinn, 1970), system pressure (Letzel et al., 1997), and temperature (Lin et al., 2001). The bubble size within the column is believed to approach a stable size shortly after dispersion, making mass transfer within the vessel less sensitive to sparger design (Deckwer and Schumpe, 1993). A stirred tank reactor is a cylindrical vessel that is equipped with an impeller at its center. Mixing and mass transfer are often enhanced by placing a sparger under the impeller to generate very small gas bubbles that have large surface area per unit volume and increase turbulence in the liquid (Shi et al., 2014). A jet loop reactor, on the other hand, is a vessel that houses a twomixture nozzle, which may be fixed at the top or bottom of the reactor. The vessel is also equipped with a draft tube that may either be concentric with the main tube or next to it (Gaddis, 1999). The momentum created by the liquid jet leads to good circulation of the gas-liquid mixture, which in turn leads to good mixing in these type of reactors with no dead zones (Zhang et al., 2014). The impinging stream reactor is a special case of the jet loop reactor, where both gas and liquid are fed to the reactor through nozzles placed at the inlet of guide tubes, forming a homogeneous twophase stream. The kinetic energy associated with the two-phase streams generates a high turbulence and, consequently, a large mass transfer area between the two phases (Gaddis and Vogelpohl, 1992). Another similar reactor is the reciprocating jet reactor, which consists of several perforated discs joined together through a central shaft. The shaft and discs are housed in a cylindrical vessel that is subjected to a counter motion with high amplitude and frequency, pushing the mixture through the perforated discs in the form of strong jets (Gaddis, 1999).

Although the above-mentioned gas-liquid contactors have been widely used in many industries, none of them can be applied to a variety of unit operations with the same efficiency, and they all seem to suffer from different drawbacks such as complexity, high demand for mechanical energy and difficulty to scale-up. The current study describes a novel system that can provide excellent gasliquid contact, high performance efficiency and can be easily scaled-up. The system is suitable for numerous gas-liquid contact applications, such as the sweetening of natural gas, solvent regeneration and CO₂ capture. The system performance and dynamic behavior were evaluated for the capture of CO₂ through reactions with ammonium hydroxide at different conditions.

2. Description of the contactor/reactor system

The system consists of a jacketed, stainless steel cylindrical vessel with 78 mm internal diameter, 700 mm height, and a total working volume of 3000 ml. The liquid enters at the top of the contactor, while the gas is injected through an orifice at the bottom of the conical base of the vessel as shown in Fig. 1. The gas leaves at the top of the vessel and the effluent liquid leaves at the same level as the liquid inlet through a tube inserted all the way to the bottom of the vessel. The liquid effluent is pushed through the effluent tube by the liquid hydrostatic pressure in the vessel. This mechanism simulates a counter-current flow, improves the gas-liquid contact and eliminates the possibility of gas entrainment with the effluent liquid. The pressure needed to force the gas through the liquid outlet tube at the bottom of the reactor is very high compared to the rest of the reactor vessel and hence no gas will be entrained with the effluent liquid. The reactor is also equipped with a fine screen at the top, which acts as a demister to prevent entrainment of liquid droplets or particles with the effluent gas. The contactor vessel is equipped with inert particles ranging in size from 5 to 15 mm and represent 5–10% of the total vessel volume. The particles must be inert in relation to the gas-liquid system and should not be reacted or affected by contact with the gas or the liquid in the vessel. The density of the particles must be very similar to that of the liquid so that they can move easily within the vessel. The particles create vigorous mixing within the vessel through systematic circular motion and provide high gas-liquid interfacial area for effective mass transfer between the two phases. The system is suitable for gas-liquid reactions, especially those involving gas solubility in the liquid followed by reaction. It can also be used for mass transfer operations involving stripping or absorption such as amine acid gas removal and amine regeneration. The size of the orifice, the gas velocity as well as the gas to liquid ratio depend on the type of the gas contact or the reaction system. The good mixing and high interfacial area are essential for operations involving absorption, stripping or gas-liquid reactions.

3. Experimental evaluation

3.1. *Gas-liquid reaction efficiency*

The contactor system was evaluated for the capture of CO_2 through reactions with ammonium hydroxide. The reaction was carried out through contacting a gas mixture containing 10% CO_2 and 90% air with ammonium hydroxide (25% NH₃) mixture with

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