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Numerical Study on Post-Combustion Technologies for CO_2 Capture

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(燃烧后二氧化碳捕捉技术流场数值模拟)

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Preface

Since last century, the global warming problem has become more and more serious. People are all looking for active solutions. Over the years, the Chinese government has been actively implementing international environmental conventions, promoting clean development mechanism, to contribute efforts to curb global warming. Scientific consensus exists on the primary driving mechanisms for global increases in atmospheric carbon dioxide (CO_2) concentrations—they are anthropogenic emissions from the use of fossil fuels and changes in land use. Hence, if the deleterious effects of rising greenhouse gas (GHG) concentrations are to be averted or avoided, an imperative need exists to reduce the extent to which they are emitted into the atmosphere.

Currently, technologies are under development for capturing and sequestering CO_2, with a primary focus on power generation. Technologies for CO_2 capture include pre-combustion, post-combustion, oxy-fuel combustion and chemical looping combustion scenarios. Post-combustion capture is removing CO_2 from the flue gas, a stage when fuel is already combusted, is the most technologically mature, and may be implemented at the large-scale needed for application to power production in the near future.

Packed bed reactors with counter-current, gas-liquid flows have been considered to be applicable in CO_2 capture systems for post-combustion processing. However, the hydrodynamics within the packing used in these reactors under counter-current flow has not been assessed to provide insight into design and operational parameters that may impact reactor and reaction efficiencies. Experimental studies of packed bed reactors and reactions are very time consuming and expensive, also, it is very difficult to measure the very complex geometry of most packed bed materials. Especially when limited space exists between the packing, it is impractical to insert measuring devices which by themselves may disturb or alter liquid and gas flow patterns. Instead of an experimental approach, computational fluid dynamics (CFD) has become a more useful tool for studying multiphase flows with complicated geometries, and its use and development may become important for reducing the number and costs of experimentation and for discovering information which otherwise may be impossible to uncover during experimentation.

This book mainly has 6 chapters to discuss the numerical study on Post-Combustion technologies for CO_2 capture.

Chapter 1 is the introduction for this book. It includes the research motivation and objectives.

Chapter 2 provides a comprehensive literature review on previous research in this field. The status of research for liquid flow regimes, distributions, holdups, and pressure drops are discussed; a status of understanding wetted surface areas of packing materials is given and a closure model in different packed beds which assesses current experimental and CFD methods and results is presented. Also gaps in data and knowledge are identified, and shortcomings are analyzed which may be overcomed by the research carried out through this book.

Chapter 3 introduces the hydrodynamic assessment on ameso-scale while using a spherical ball packed bed reactor. The outcome of the CFD model will be compared to data like pressure drops from experimentation that was accomplished while using various mass flow conditions. This hydrodynamic assessment also examines relationships between the flow regime, flow distributions, pressure drops, liquid holdups, wetted areas and the operating conditions.

Chapter 4 introduces the hydrodynamic study on ameso-scale model on a structured packed bed reactor. The numerical model is validated by comparing simulated and empirical pressure drop model from scientific literature. Also the author analyzed the relationships between flow regimes, flow distributions, pressure drops, liquid holdups, wetted areas and the operating conditions.

Chapter 5 presents the hydrodynamic study using a macro-scale model and a closure model from literature data. A new closure model was modified.

Chapter 6 is conclusion and future work.

It has been a long journey to complete this book, and fortunately my advisor Kunlei Liu and Dr. Kozo Saito have provided a lot of help. I wish to express my gratitude to them. And also thank for the financial support from the Key Laboratory of Coal-based CO_2 Capture and Geological Storage of China University of Mining and Technology (CUMT)、the Natural Science Foundation of Jiangsu Province and the support from 2018 Double-first-class Construction of Independent Innovation project of CUMT.

Author
Xuzhou, August 15, 2018

Contents

Chapter 1 Intoduction

1.1 Research Motivation

Scientific consensus exists on the primary driving mechanisms for global increases in atmospheric carbon dioxide (CO_2) concentrations—they are anthropogenic emissions from the use of fossil fuels and changes in land use. The use of coal alone accounted for 43% of the total global CO_2 emissions in 2010[1] and is a part of the reason that atmospheric CO_2 concentrations climbed to 403.7 ppm in May, 2015[2], a value 40% above pre-industrial levels. Other greenhouse gases (GHG), including methane (CH_4), water vapor (H_2O) and ozone (O_3), are also recognized as the main atmospheric constituents that affect global warming trends. Hence, if the deleterious effects of rising GHG concentrations are to be averted or avoided, an imperative need exists to reduce the extent to which they are emitted into the atmosphere [3].

Currently, technologies are under development for capturing and sequestering CO_2, with a primary focus on power generation. Technologies for CO_2 capture include pre-combustion, post-combustion, oxy-fuel combustion and chemical looping combustion scenarios. Post-combustion capture is removing CO_2 from the flue gas, a stage when fuel is already combusted, and solvent-based CO_2 absorption is one of the most popular approaches [4-5]. Pre-combustion capture is the removal of CO_2 from syngas prior to its combustion, usually in application with Integrated Gasification Combined Cycle (IGCC)[6-7]. Oxy-fuel combustion uses pure oxygen to combust fuels instead of air, so nitrogen is not involved in the combustion process. Thus CO_2 in the flue gas is in high concentration [8-9]. Chemical looping combustion belongs to the category of oxy-fuel combustion. It uses oxygen carriers to provide oxygen instead of a costly air separation unit (ASU), thus the energy penalty is low [3,10-16]. Of these carbon capture technologies, post combustion CO_2 capture is the most technologically mature, and may be implemented at the large-scale needed for application to power production in the near future.

1.1.1 Amine-based Post Combustion CO_2 Capture

A process diagram of a post-combustion CO_2 capture using a packed bed reactor is presented in Figure 1-1 [17-18]: it includes two reactors, one of which is called an absorber and the other is a stripper. An aqueous solution that is usually amine based circulates between the two reactors in which one absorbs CO_2 (absorber) and the other releases CO_2 (stripper) to produce a flow of highly concentrated CO_2. In the absorber, the CO_2 and solvent are in counter-current flow where CO_2 is absorbed by the amine-based solvent, thereby producing a gas outlet product that is CO_2-depleted; simultaneously, the CO_2 enriched amine exits the column from the bottom of the absorber and flows into the top of the stripper. In the stripper, the CO_2 enriched amine solution is heated up to a temperature exceeding 100 °C to release CO_2 and generate a high purity CO_2 stream that is ready for sequestration. The regenerated amine (without CO_2) is pumped back to the top of the absorber for a next cycle. Because the amine from the stripper is at a relatively high temperature, a heat exchanger (HEX) is typically applied for improving overall process efficiency.

The absorber is a packed bed reactor which is filled with packing materials. The main objective of using a packed bed is to improve the effective contact between the gas and liquid phase reactants.

1.1.2 Packed Bed Reactors

The packed bed reactor, also called a trickle bed reactor, was originally designed for wastewater treatment but has become very useful in diverse

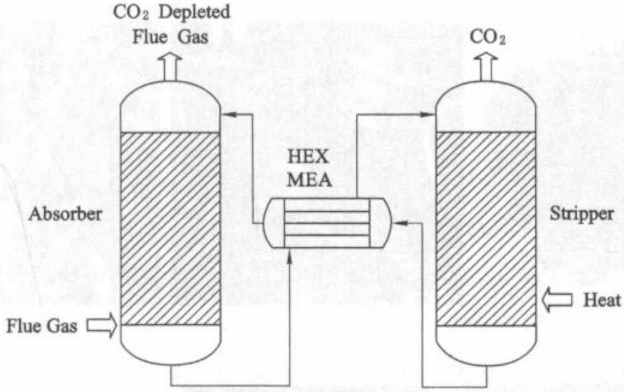

Figure 1-1 Post combustion solvent based CO_2 capture process

industries in which two-phase flow and chemical reactions are prevalent. In general, a packed bed is a hollow tube, pipe or other vessel that is filled with packing materials[19]. The packing materials can be small objects like spherical balls, rings or specifically designed, structured packing materials, arranged in random or structured orientations. A random packing consists of randomly distributed packing material within the bed, while structured packing consists of uniformly arranged material in the bed. Figure 1-2 shows some examples for random packing and structured packing.

Packing materials improve the contact area at which chemical or physical reactions take place and, thereby, can increase markedly the efficiency by using a packed bed reactor. As an example, for gas-liquid multi-phase flow in a packed bed, the liquids will spread onto the packing materials and tend to wet its surface to form a liquid film layer; then when a gas flows across the packing material, the gas-liquid contact area within the reactor will therefore be enhanced. This contact area is where mass transfer occurs[20], and increasing contact area is one important pathway recognized to enhance mass transfer and chemical reactions. Therefore, the use of packing bed promotes reactor and reaction advantages such as high capacity, high efficiency and, if designed appropriately, low pressure drop.

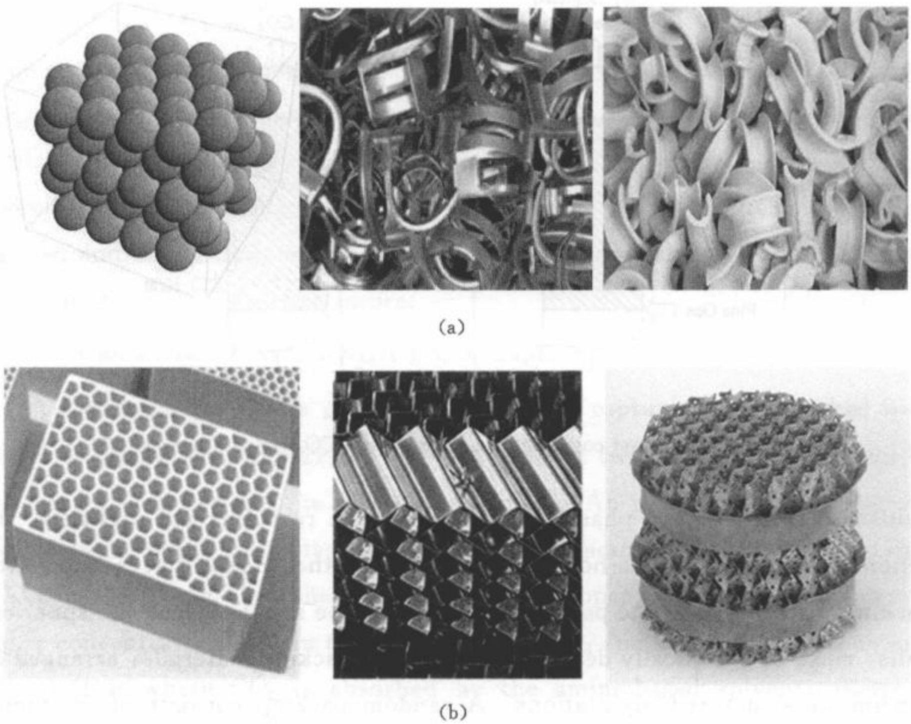

Figure 1-2 Packed beds with (a) random and (b) structured packing materials

Different-shaped packing materials provide possibilities to vary contact areas and void space between the packing. Both of these factors affect performance, but typical selection of a packing material depends, to a large extent, on the application and the cost. In general, random packing is a lower cost approach as compared to structured packing, and this advantage leads random packed beds to be used most frequently during the next decades [21]. However, structured packing can provide higher separation efficiency, higher capacity and better radial mixing than random packing. Due to these advantages, it has been estimated that 25% of all refinery vacuum towers are now equipped with structured packing[20]. Because both random and structured packing are used throughout industry but their assessment and application within CO_2 capture technological applications are only beginning, it is important to study the hydrodynamics like liquid distribution, hold-up and wetted area caused by the packing materials to enable more accurate and efficient reactor designs that will be needed to minimize investment and operation costs of the huge systems envisioned for the power industry [22]. For example, for a CO_2 absorber capable of handling emissions from only a 0.7 MW power system, a height of approximately 19.5 m would be required with an inside packing material covering a length at least 13.7 m. However, a commercial CO_2 capture capacity would be typically near and greater than 300 MW, a scale-up of greater than 300 times. Hence, improving CO_2 capture rates is a decisive way to limit the size of commercial reactors, and achieving this goal can be assisted by understanding the hydrodynamics inside a packed bed where gas-liquid interfaces control the important reactions.

Although the most mature technology for CO_2 capture is in a post combustion operational mode, all CO_2 capture technological options are still in their infancy because no full-scale commercial system on an actual fossil fuel power plant has ever been built. A schematic of a packed bed reactor with spherical ball packing materials is shown in Figure 1-3. The reactants, i. e. amine solution and CO_2-laden gas flow counter-currently across the packing.

Since packed bed reactors are widely used for gas-liquid flow reactions, and the packing is one of the key factors affecting overall reaction rates. Therefore, understanding the hydrodynamics within packed beds is essential. Although extensive investigations have been performed on packed bed reactors over the years, the current understanding of flows within them is insufficient to enable optimization within the large scales needed for applications within power production[23,24]. Furthermore, experimental studies of packed bed reactors and reactions are very time consuming and expensive, not only because of extensive scale-up needs for transition from a laboratory to a commercial system but also because of the high number of options that are possible for the interior components, or packing, of the reactors. Also,

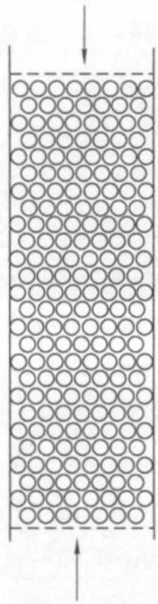

Figure 1-3 A schematic of a fixed bed reactor

experimentally it is very difficult to measure the very complex geometry of most packed bed materials and, especially when limited space exists between the packings, it is impractical to insert measuring devices which by themselves may disturb or alter liquid and gas flow patterns.

Hence, this book focuses on packed bed reactors and packing for application to study multiphase flow and hydrodynamics during post combustion CO_2 capture. Instead of an experimental approach, computational fluid dynamics (CFD) is the primary tool used during the research[25]; when possible, the results of the CFD studies are compared to laboratory-scale experimentation. Scale modeling technique[26,27] offers basic concepts and methods to use scale model experiments to validate numerical modeling predictions. Figure 1-4 shows the concept of scale and numerical modeling methods[28]. After this validation is made, numerical modeling approach leverages the ever-increasing computational power of computers with sophisticated CFD software. Because the geometry of any one packing material can be very complex and the resulting flow patterns also demand to be simulated, a need exists to focus on a limited number of predetermined packing configurations that are described and discussed in the following chapters.

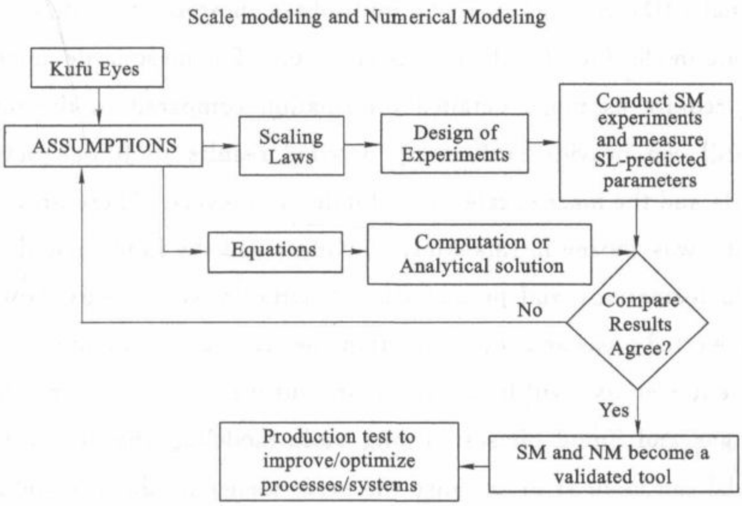

Figure 1-4 Basic concepts of scale modeling and numerical simulation [28]

Historically, the size of reactors and the number of packing elements under assessment have been limited by computational constraints. It has been a challenge to simulate the whole domain of a reactor with its packing material but symmetry, selected arrangements of the packing structures and judicious meshing choices make CFD assessments more productive than pure experimentation. To date, some computational research has been performed based on a meso-scale model approach, however, these simulations have been based on single phase, or gas-liquid phase in co-current flow [24,29-41]. What is missing is an accurate assessment on multiphase and counter-current flow system, a current trend in the packed bed reactors applied for post combustion CO_2 capture. This book, therefore, focuses on this CO_2 capturing system.

1.2 Objectives

The overall goal of this book is to develop: (1) a comprehensive three dimensional (3D) counter-current, multiphase meso-scale model, and (2) a macro-scale model for CO_2 absorber simulation. The meso-scale model, which does not require as much detailed information compared to the micro-scale model, still can provide sufficiently detailed results to bridge between the micro-scale and the macro-scale (actual industrial level). Therefore, the meso-scale model was chosen in this study to understand the fundamental aspects of liquid film formation, and predict flow distributions, pressure drops, liquid holdups, wetted areas and the interactions between gas and liquid phases. The meso-scale model also will be used to point out ways for enhancing the contact between gas and liquid phases. Using scale modeling theory[32], the meso-scale model can be used to optimize the performance of macro-scale model.

Specific objectives to achieve the aforementioned goal are to:

(1) develop a comprehensive 3D CFD model for simulating liquid-gas counter-current multiphase flow while using both spherical ball and structured packing, and then to verify the model with experimental data;

(2) obtain insight into film formation, flow distributions, pressure drops, liquid holdups, wetted areas and the interactions between phases in both spherical ball and structured packing;

(3) assess the effects of operating conditions on film formation, flow distributions, pressure drops, liquid holdups, wetted areas and the interactions between phases and then to develop a working relationship between these parameters and the operating conditions;

(4) develop a fundamental understanding of controlling factors influencing gas-liquid interactions; and

(5) modify and optimize a macro-scale packed bed model for simulating the hydrodynamics in a CO_2 capture process using amine-based liquids.

However, this book does not offer detailed analysis and discussions on rigorous scaling relationships and scaling laws that can relate all three scales: mico-scale, meso-scale and macro-scale. This scaling study is important and interesting, but simply beyond the scope of this monograph and therefore leaves it for future studies.