Lunbo Duan
Dennis Lu

Fluidized Bed Reactors for Carbon Capture

A Review of Advancing Combustion and Sorption Techniques for Decarbonization

OPEN ACCESS

Springer

Fluidized Bed Reactors for Carbon Capture

Lunbo Duan · Dennis Lu

Fluidized Bed Reactors for Carbon Capture

A Review of Advancing Combustion and Sorption Techniques for Decarbonization

Springer

Lunbo Duan
Key Laboratory of Energy Thermal Conversion and Control of Ministry of Education
School of Energy and Environment
Southeast University
Nanjing, Jiangsu, China

Dennis Lu
Key Laboratory of Energy Thermal Conversion and Control of Ministry of Education
School of Energy and Environment
Southeast University
Nanjing, Jiangsu, China

ISBN 978-981-96-0273-5 ISBN 978-981-96-0274-2 (eBook)
https://doi.org/10.1007/978-981-96-0274-2

This work was supported by Dennis Yong Lu.

© The Editor(s) (if applicable) and The Author(s) 2025. This book is an open access publication.

Open Access This book is licensed under the terms of the Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/), which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license and indicate if changes were made.
The images or other third party material in this book are included in the book's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the book's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.
The use of general descriptive names, registered names, trademarks, service marks, etc. in this publication does not imply, even in the absence of a specific statement, that such names are exempt from the relevant protective laws and regulations and therefore free for general use.
The publisher, the authors and the editors are safe to assume that the advice and information in this book are believed to be true and accurate at the date of publication. Neither the publisher nor the authors or the editors give a warranty, expressed or implied, with respect to the material contained herein or for any errors or omissions that may have been made. The publisher remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

This Springer imprint is published by the registered company Springer Nature Singapore Pte Ltd.
The registered company address is: 152 Beach Road, #21-01/04 Gateway East, Singapore 189721, Singapore

If disposing of this product, please recycle the paper.

Preface

In the escalating battle against global warming, Carbon Capture, Utilization, and Storage (CCUS) technologies have emerged as a beacon of hope. As industries and nations alike strive towards a carbon-neutral future, the role of CCUS in decarbonizing industrial production and mitigating climate change is undeniably pivotal. Among the various technologies explored for effective carbon capture, the application of fluidized bed reactors (FBRs) holds significant promise due to their efficiency and adaptability in large-scale operations. This book presents an in-depth exploration of fluidized bed technologies, focusing on their role in CO_2 capture, and is designed to serve as a comprehensive resource for both academic research and industrial applications.

The inception of this book is rooted in the collective realization of the urgent need for advanced solutions to tackle CO_2 emissions effectively. With global temperatures rising and the detrimental effects of climate change becoming increasingly evident, there is a pressing need to develop and implement technologies that can reduce atmospheric CO_2 levels. CCUS technologies have been recognized as crucial in this endeavor. This book meticulously reviews a suite of CO_2 capture technologies that employ fluidized bed reactors, including pre-combustion gasification, oxyfuel combustion, chemical looping combustion, calcium looping processes, and low-temperature sorption. Our primary objective is to provide a thorough and comprehensive examination of fluidized bed reactors in the context of CO_2 capture. This involves summarizing the state-of-the-art progress from lab-scale fluidized bed reactors to pilot-scale demonstration facilities, discussing the remaining challenges in the field, and offering insights into potential solutions and future advancements. By doing so, we aim to pave the way for the widespread commercialization of these technologies.

Our coverage begins with a foundational overview in the introductory chapter, setting the stage for a deeper exploration of the operational fundamentals of fluidized bed reactors. Detailed discussions on the design, operation, and critical aspects of heat and mass transfer within FBRs are provided to elucidate their relevance to carbon capture efficiency. The book progresses into specific technologies, starting with gasification in FBRs, exploring recent advancements and design challenges, and culminating in detailed case studies and performance evaluations. The subsequent chapters are dedicated to oxy-fuel combustion, chemical looping combustion, and calcium looping, each dissecting the technological developments, operational insights, and economic viability of these processes. In particular, the discussion on chemical looping combustion highlights the selection and performance of oxygen carrier materials, a core component that significantly influences the efficiency of the process. The penultimate sections of the book explore the innovative realm of low-temperature sorption in FBRs, focusing on sorbent development and process optimization, before scaling up to demonstration projects that showcase the practical viability of these technologies. Each chapter concludes with a reflection on the challenges faced and prospective solutions that hold the potential to overcome these barriers. In the final chapters, we address the broader challenges and outlook on the integration of these technologies into existing industrial frameworks, emphasizing the technical hurdles, economic barriers, and need for innovation in future research priorities.

We would like to express our deepest gratitude to a remarkable group of experts who generously shared their profound insights and invaluable suggestions during the preparation of this book. We are particularly grateful to Prof. Bo Leckner of Chalmers University of Technology, Sweden, who meticulously reviewed all chapters and provided us with useful feedback in both general and specific areas. We also extend our appreciation to Prof. Zhenkun Sun and Drs. Hongjian Tang, and Yuanqiang Duan, as well as Yueming Wang of Southeast University, China, for their contributions in drafting and editing several chapters in this book. We acknowledge the assistance provided by students Lu Chen, Caili Li, and Tongxing Zhang, who helped us obtain permission to use charts and tables from other publishers. Lastly, we would like to express our gratitude to our colleagues, peers, and the wider academic community for their invaluable contributions, which have significantly enhanced the content and scope of this book. This book owes its publication to the generous support of the National Natural Science Foundation of China (No. U22A20435) and the Start-up Research Fund of Southeast University (RF1028623358). We extend our heartfelt thanks to these organizations for their financial support, which has been critical to the completion of this work.

The culmination of this book not only synthesizes the current knowledge but also outlines a future trajectory for research and development in fluidized bed technologies for CO_2 capture. We hope that this comprehensive review acts as a catalyst for further studies and technological advancements in this critical field. As we present this book to the readers, we hope that it serves as a valuable resource and inspires continued research and innovation in the field of carbon capture using fluidized bed technologies. We are excited about future possibilities and eager to see how the next generation of researchers and practitioners will take this knowledge forward and contribute to a sustainable and carbon-neutral future.

Nanjing, China
May 2024

Prof. Lunbo Duan
Prof. Dennis Lu

Contents

Chapter 1
Introduction

Climate change is currently one of the most challenging issues to the global ecosystem, which has already caused serious consequences in many aspects, such as increasing wildfires, heat waves, drought, and ice sheet disintegration [1]. Anthropogenic CO_2 emission, mainly produced by power generation and industrial processes, has been well-documented as the primary cause of existing global warming, accounting for ~76% of total greenhouse gas emissions [2]. The extensive use of fossil fuels has significantly increased atmospheric CO_2 concentration over the past centuries [3]. Hence, with the global ambition to limit the rise of global average temperature preferably to 1.5 °C and to achieve net-zero emissions by 2050, many countries and regions worldwide have proposed clear guidelines towards carbon neutrality around the second half of this century [4].

While there is a global consensus that CO_2 emission control is pivotal to mitigating climate change, the challenge lies in finding economically and practically viable ways to reduce CO_2 emissions. For example, developing countries are likely to continue relying on fossil fuels in the near future because of their availability, reliability, and affordability. However, it is worth noting that the power generation sector accounts for approximately 38.8% of global CO_2 emissions [5]. Therefore, it is crucial to prioritize the adoption of technically viable measures to capture CO_2 from fossil fuel combustion, particularly in the context of energy security [6]. Carbon capture technologies can be mainly divided into pre-combustion, oxy-fuel combustion, and post-combustion routes [7]. After capturing CO_2, it must be purified to eliminate impurities, often using separation technologies like membranes or solvents. The purified CO_2 is then compressed for easier transportation and storage, it can be transported via pipelines, trucks, or ships, with stringent safety measures to prevent leaks. Its utilization includes enhanced oil recovery [8], conversion into fuels [9], or other chemical products [10]. These methods can transform CO_2 from waste to a valuable resource, promoting a circular carbon economy. Among these, this paper mainly focuses on carbon capture technologies.

© The Author(s) 2025

L. Duan and D. Lu, *Fluidized Bed Reactors for Carbon Capture*,
https://doi.org/10.1007/978-981-96-0274-2_1

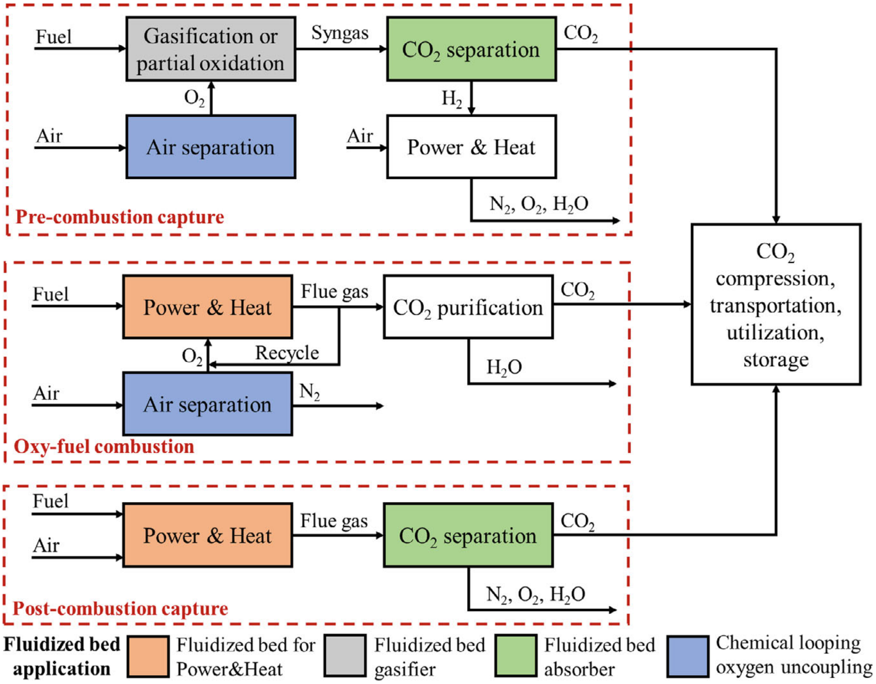


Fig. 1.1 Potential applications of FBRs for pre-combustion, oxyfuel combustion, and post-combustion carbon capture technologies

Technically, the selection of appropriate reactor configuration is a shared challenge for the above-mentioned carbon capture technologies, especially as the CO_2-sorbents contact is tightly linked to the CO_2 capture efficiency [11]. Fluidization is a physical phenomenon that occurs when a solid particulate substance inside the reactor behaves in a fluid-like characteristic. The operation of a fluidized bed reactor (FBR) involves injecting the gaseous media at a high velocity through the bottom of the reactor to flow through the bed materials. The fluidization characteristic makes the FBR more promising in terms of gas-solid mixing, heat transfer rate, fuel adaptability, operation flexibility, and pollutant control. Consequently, fluidized beds find extensive applications across diverse carbon capture technologies, as illustrated in Fig. 1.1.

The publication about the fluidized bed application in these areas has significantly increased in recent years (Fig. 1.2). However, FBRs face several technical barriers that hinder their industrial application in carbon capture technologies due to existing technical barriers, e.g., high energy penalties [11], scale-up issues for commercialization [12], and selection of suitable high-reactivity materials [13]. More technical advancements are now under investigation to further improve the system's efficiency and flexibility, such as the integration with renewable energy and energy storage [14–16]. Therefore, this review aims to scrutinize the pivotal role of FBRs in the

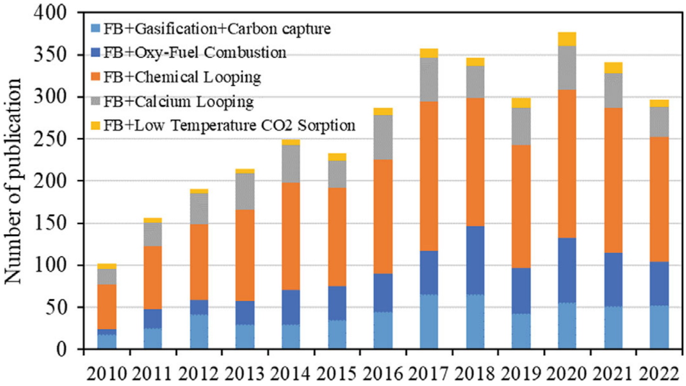


Fig. 1.2 Development trend of literature concerning fluidized bed's application in CCUS from 2010 to 2022. *Data source* Web of Science (WOS) core collection, and here only include articles and reviews

arena of carbon capture technologies. A critical evaluation of current literature, as well as the assessment of technological advancements, is undertaken to understand the spectrum of operational dynamics, material innovations, and the thermodynamic proficiency of FBRs in carbon capture applications. In addition to scrutinizing the role of FBRs in carbon capture, this review also aims to provide a critical analysis of the latest technological advancements and innovative applications within this domain. It includes an examination of emerging trends in FBR design, operational efficiency, and adaptability to varying carbon capture methods. The scope extends to evaluating a comparative assessment of FBRs against other carbon capture technologies, highlighting their unique advantages and limitations, thereby providing a comprehensive overview that is relevant to industry practitioners and the academic community.

By dissecting the latest innovations and identifying operational challenges, the review sets out to offer potential solutions and project future directions for research and development. The ultimate goal is to furnish a detailed synthesis of FBR technology, assessing its current state and future prospects within the broader context of environmental sustainability and economic viability. It aims to provide actionable insights and strategic recommendations for advancing FBR technology, catering to the needs of researchers, industry experts, and policymakers.

The structural outline of the review is methodically crafted to facilitate a coherent understanding of FBRs. Following an introductory exposition, the review will progress into a detailed discussion of the operational fundamentals of FBRs, laying the groundwork for understanding their application in various carbon capture processes. Subsequent sections are dedicated to an in-depth exploration of specific carbon capture methods, such as oxy-fuel combustion, chemical looping combustion (CLC), and the calcium looping process (CaL). The discourse extends to exploring the integration of low-temperature sorption processes within the FBR framework, highlighting the innovative use of novel sorbents. Each of these sections will not only discuss the technical details but also critically analyze the performance, cost implications, and environmental impact. The review will then transition into an evaluation of recent innovations and challenges, drawing insights from current research and industrial practices. This will include a discussion on the development of novel materials and processes, as well as an examination of operational hurdles and potential solutions. In the same section, the review will synthesize the findings to offer a comprehensive outlook on the future of FBRs in carbon capture. The objective is to foster a deeper understanding of the role of FBRs in addressing climate change challenges and to stimulate further research and innovation in this field.

References

1. Masson-Delmotte V, Zhai P, Pörtner H-O, Roberts D, Skea J, Shukla PR et al (2018) Global warming of 1.5 ºC. An IPCC Special Report on the impacts of global warming of 15 ºC 1
2. Stocker T (2014) Climate change 2013: the physical science basis: working group I contribution to the fifth assessment report of the intergovernmental panel on climate change. Cambridge University Press
3. Wuebbles DJ, Jain AK (2001) Concerns about climate change and the role of fossil fuel use. Fuel Process Technol 71:99–119
4. Wei Y-M, Han R, Wang C, Yu B, Liang Q-M, Yuan X-C et al (2020) Self-preservation strategy for approaching global warming targets in the post-paris agreement era. Nat Commun 11:1–13
5. Yoro KO, Daramola MO (2020) Chapter 1—CO_2 emission sources, greenhouse gases, and the global warming effect. Elsevier
6. Herzog H, Golomb D (2004) Carbon capture and storage from fossil fuel use. Encycl Energy 1:277–287
7. Rubin ES, Mantripragada H, Marks A, Versteeg P, Kitchin J (2012) The outlook for improved carbon capture technology. Prog Energy Combust Sci 38:630–671
8. Farajzadeh R, Eftekhari AA, Dafnomilis G, Lake L, Bruining J (2020) On the sustainability of CO_2 storage through CO_2–enhanced oil recovery. Appl Energy 261:114467
9. Wei J, Ge Q, Yao R, Wen Z, Fang C, Guo L et al (2017) Directly converting CO_2 into a gasoline fuel. Nat Commun 8:15174
10. Saravanan A, Vo D-VN, Jeevanantham S, Bhuvaneswari V, Narayanan VA, Yaashikaa P et al (2021) A comprehensive review on different approaches for CO_2 utilization and conversion pathways. Chem Eng Sci 236:116515
11. Dhoke C, Zaabout A, Cloete S, Amini S (2021) Review on reactor configurations for adsorption-based CO_2 capture. Ind Eng Chem Res 60:3779–3798
12. Seddighi S, Clough PT, Anthony EJ, Hughes RW, Lu P (2018) Scale-up challenges and opportunities for carbon capture by oxy-fuel circulating fluidized beds. Appl Energy 232:527–542
13. Raganati F, Miccio F, Ammendola P (2021) Adsorption of carbon dioxide for post-combustion capture: a review. Energy Fuels 35:12845–12868
14. Di Lauro F, Tregambi C, Montagnaro F, Salatino P, Chirone R, Solimene R (2021) Improving the performance of calcium looping for solar thermochemical energy storage and CO_2 capture. Fuel 298:120791
15. Ortiz C, Valverde JM, Chacartegui R, Pérez-Maqueda LA, Gimenez-Gavarrell P (2021) Scaling-up the calcium-looping process for CO_2 capture and energy storage. Kona Powder Part J 38:189–208
16. Saghafifar M, Gabra S (2020) A critical overview of solar assisted carbon capture systems: is solar always the solution? Int J Greenhouse Gas Control 92:102852

Open Access This chapter is licensed under the terms of the Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/), which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license and indicate if changes were made.

The images or other third party material in this chapter are included in the chapter's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the chapter's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.

Chapter 2
Fluidized Bed Reactors: Operational Fundamentals

2.1 Introduction to Fluid Dynamics in FBRs

Fluidization facilitates a dynamic interplay between solid particles and upward-flowing gases, essential to a multitude of industrial processes such as drying [1], coating [2], granulation [3], casting [4], pyrolysis [5], gasification [6], and energy storage [7]. At the core of fluidization is the suspension of solid particles in a state akin to a fluid, achieved by passing a gas upward through a bed of solid particles with sufficient velocity to exert drag forces that counterbalance the particles' weight [8]. This fluid-like state is not a mere technical novelty; it is the linchpin of effective gas–solid interactions, thermal conduction, and chemical reactivity. It provides an optimal milieu for diverse chemical reactions, including those pivotal in carbon capture technologies.

The dynamics of particles within an FBR are governed by their intrinsic properties—namely size, density, and shape—as well as the properties of the fluidizing medium, such as velocity and density. Grasping these dynamics is imperative for refining reactor design and operational protocols, particularly in carbon capture endeavors. The Geldart classification system offers a structured approach to categorizing powders based on their aerodynamic characteristics, which are instrumental in determining their fluidization profile (refer to Fig. 2.1) [9]. Within this system, particles are segmented into four principal categories [10]:

- Group A particles are diminutive and cohesive, prone to uniform fluidization, usually under 30 microns in diameter. They necessitate meticulous control over air velocity to avert channeling and secure a uniformly fluidized bed.
- Group B particles, with diameters ranging from 40 to 500 microns, are often deemed optimal for fluidization due to their ease of fluidization and minimal inter-particle cohesion, rendering them versatile for a broad spectrum of FBR applications.

© The Author(s) 2025

L. Duan and D. Lu, *Fluidized Bed Reactors for Carbon Capture*,
https://doi.org/10.1007/978-981-96-0274-2_2

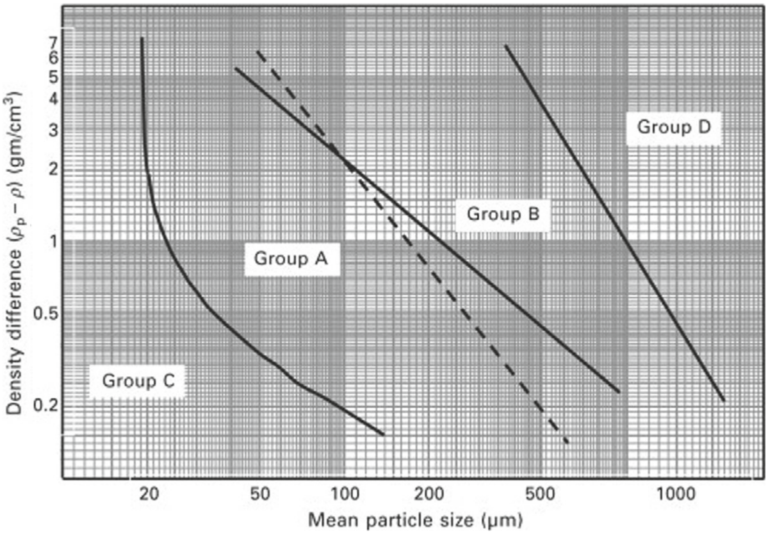


Fig. 2.1 Geldart classification of particles [9]

- Group C encompasses fine, cohesive particles that pose fluidization challenges and may require auxiliary measures such as mechanical agitation to realize fluidization.
- Group D contains larger, denser particles that are inclined towards spouted or turbulent fluidization, demanding higher velocities to become fluidized.

In reactor design for carbon capture, the chosen fluidization regime is pivotal, see Fig. 2.2, as each regime-bubbling, circulating, turbulent, or fast fluidization—engenders distinct fluid dynamics and transport phenomena, impacting efficiency [11].

- Bubbling Fluidization: Characterized by lower gas velocities, this regime leads to bubble formation within the bed. While these bubbles enhance gas–solid mixing, they can simultaneously detract from the efficiency of contact between the reactant gas and solid sorbents.
- Circulating Fluidization: Operating at higher velocities, the circulating fluidized bed (CFB) regime entrains solid particles out of the bed. These particles are then separated from the exit gas and recirculated into the reactor. This continuous loop endows CFB reactors with excellent gas–solid contact and heat transfer capabilities, making them highly suitable for carbon capture.
- Turbulent and Fast Fluidization: At even higher gas velocities, the bed transitions into a turbulent state, with vigorous particle movement and intense mixing—known as fast fluidization. These regimes are advantageous for their augmented mass and heat transfer rates, which can significantly enhance the kinetics of carbon capture reactions.

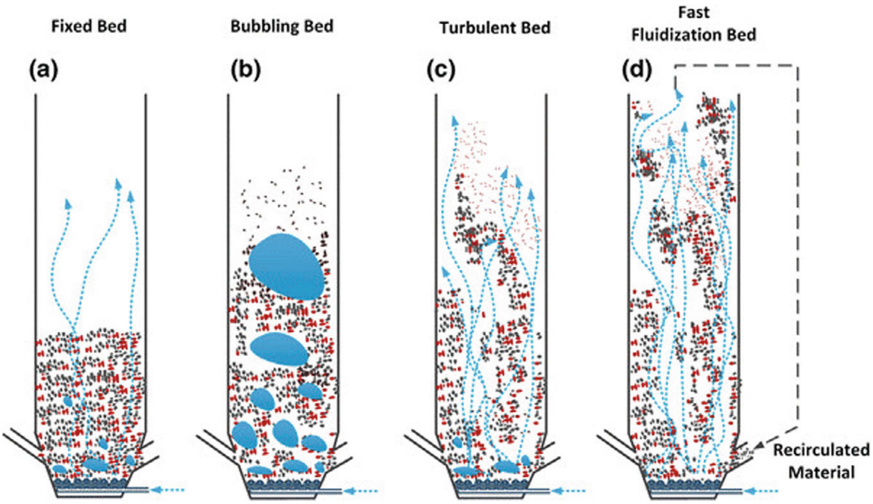


Fig. 2.2 Categories of fluidization regimes [11]

Comprehension of fluidization principles and the associated regimes is imperative for the enhancement of FBR efficacy in carbon capture applications. The meticulous regulation of fluidization parameters, including gas velocity, particle size distribution, and bed height, is crucial [12]. Such control is necessary not only to augment CO_2 absorption efficiency but also to curtail energy demands and guarantee stable, enduring operation. This introduction lays the groundwork for an in-depth examination of FBR design and operational strategies, steering us toward the development of scalable, cost-effective, and environmentally sound responses to the pressing imperative of carbon capture.

2.2 Design and Operation of FBRs

FBRs are sophisticatedly engineered systems that harness fluidization phenomena to enable a spectrum of chemical reactions, notably those pertinent to carbon capture [13]. The strategic design and astute operation of FBRs are integral to bolstering their performance and tailoring them to meet the specialized requirements of carbon capture processes.

Within an FBR, the fluidization phenomenon is dictated by the core principles of fluid mechanics [8]. When a gas ascends through a bed of solid particles at a low velocity, it traverses the interstices with minimal particle interaction. This state, reminiscent of a fluid's behavior, is indispensable for efficacious gas–solid contact, thermal exchange, and mass transfer—all of which are vital for the efficacious absorption of CO_2 from various gas streams.

2.2.1 FBR Design Features

The efficacy of an FBR is significantly contingent on its design characteristics [14]:

- Distributor Plate Design: Serving as the foundation of the FBR, the distributor plate is instrumental in introducing the fluidizing medium. Its design is paramount in ensuring an even gas spread, crucial for uniform fluidization. The plate's perforations or nozzles must be strategically dimensioned and arrayed to thwart particle backflow and diminish inactive zones within the reactor.
- Bed Material Selection: Selecting an appropriate bed material is essential, not only for facilitating fluidization but also for influencing reaction kinetics. The material's granular size, density, and surface properties must be fine-tuned for optimal interaction with the gas flow. In carbon capture, the sorbent's reactivity and ability to be regenerated are also key, as these factors directly impact the CO_2 capture efficacy.
- Reactor Geometry: The reactor's configuration, encompassing its shape and dimensions, affects fluid dynamics and the retention time of gas and solids within the system. Vertically elongated reactors may promote prolonged contact times, albeit at the potential cost of uniform gas distribution and solid recirculation. On the other hand, reactors with a broader girth may improve gas dispersal but might necessitate increased velocities for fluidization.

2.2.2 Impact on Reactor Performance

The operational proficiency of an FBR is intrinsically linked to its design intricacies [14]:

- Gas–Solid Contact: Optimal gas–solid interaction is paramount for efficient reaction kinetics. The distributor plate's architecture, coupled with the selection of the bed material, is crucial in enhancing this interaction, thereby augmenting the CO_2 absorption capability of the reactor.
- Heat Transfer: In an FBR, heat transfer efficiency is crucial, particularly for reactions that are exothermic or endothermic, as is common in carbon capture processes. Fluidization promotes elevated rates of heat exchange due to the perpetual motion and intermingling of particles, which aids in sustaining uniform temperature conditions within the reactor.
- Managing Diverse Reaction Rates: An FBR's capacity to accommodate varying reaction velocities stems from its design—especially its geometry and the conditions under which fluidization occurs. By adjusting these design parameters, FBRs can be fine-tuned to align with the specific reaction kinetics of CO_2 capture processes.

To encapsulate, the architecture and operational strategies of FBRs are central to their utility in carbon capture applications. A thorough grasp and refinement of fluidization dynamics, distributor plate configuration, bed material properties, and reactor geometry enable engineers to significantly advance the effectiveness and efficiency of FBRs in large-scale carbon capture efforts [15].

2.3 Heat and Mass Transfer in FBRs

The efficacy of carbon capture methodologies employing FBRs hinges critically on the internal mechanisms governing heat and mass transfer. These mechanisms are indispensable to the reactor's operational performance, as they have a direct bearing on reaction kinetics, the regeneration of sorbents, and the overarching efficiency of the system [16].

2.3.1 Mechanisms of Heat Transfer

In FBRs, heat transfer transpires predominantly through conduction, convection, and radiation, with convection playing the leading role due to the vigorous mixing of the fluidized particles. This intense particle movement bolsters the effective thermal conductivity within the FBR, as it ensures the continuous renewal of contact surfaces between the particles and the fluidizing medium [17].

The heat transfer coefficient (*h*) in fluidized beds is quantifiable by empirical formulas that incorporate the characteristics of the particles, the properties of the fluid, and the fluidization velocity. An exemplar of such correlations can be expressed as Eq. 2.1 [18]:

$$h = \frac{k_g}{d_p} \cdot Nu \tag{2.1}$$

where k_g is the thermal conductivity of the gas, d_p is the particle diameter, and Nu is the Nusselt number—a dimensionless quantity that can be further delineated in terms of the system's Reynolds (Re) and Prandtl (Pr) numbers.

2.3.2 Mechanisms of Mass Transfer

Mass transfer within FBRs is principally propelled by diffusion and enhanced by convection, a result of the fluidization process which ensures persistent exposure of solid particles to the gas phase, facilitating efficient mass exchange [16]. This mass transfer rate is particularly crucial in carbon capture technology as it governs the velocity at which CO_2 is adsorbed onto the sorbent particles.

The mass transfer coefficient (k_m) is calculable through the Eq. 2.2 [19]:

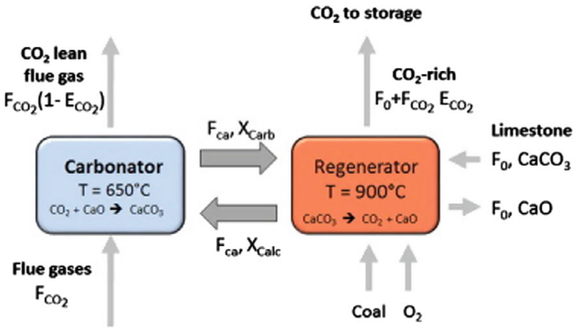


Fig. 2.3 Schematic of calcium looping with temperature requirement at each reactor (reprinted from ref. [21] with permission of Elsevier)

$$k_m = \frac{D_g}{d_p} \cdot Sh \tag{2.2}$$

where D_g is the diffusion coefficient of the gas in the solid particles, and Sh is the Sherwood number—a dimensionless entity that, akin to the Nusselt number, is dependent on the system's Reynolds (Re) and Schmidt (Sc) numbers.

2.3.3 Importance of Mixing and Temperature Control

Optimal mixing within the reactor is a cornerstone for effective heat and mass transfer, serving to homogenize temperature distributions and ensure uniform exposure of sorbents to CO_2-enriched gases—factors that are imperative for maintaining consistent reaction kinetics across the bed [20]. The modulation of mixing is orchestrated through careful manipulation of the fluidization velocity and thoughtful reactor design, which may include the strategic placement of internals such as baffles or draft tubes.

Concurrently, precise temperature regulation is of the essence as shown in Fig. 2.3, especially for thermally sensitive reactions. In the realm of carbon capture, sorbent regeneration—a thermally governed process—often demands elevated temperatures. Mastery over the reactor's thermal profile is crucial; any shortcomings in temperature regulation can precipitate incomplete sorbent regeneration, thereby diminishing the sorbent's effectiveness for CO_2 capture in ensuing cycles [21].

2.3.4 Relevance to Carbon Capture Efficiency

Temperature and concentration gradients within an FBR can be analytically delineated and forecasted through differential equations that encapsulate the principles of energy and mass conservation. To resolve these equations and derive the thermal and concentration profiles under specific operational conditions, numerical methods are frequently employed [8].

The interplay of heat and mass transfer processes within an FBR is decisive in the context of carbon capture efficiency. Proficient heat transfer is indispensable for the endothermic process of CO_2 sorbent regeneration, ensuring thorough preparation of the sorbent for successive absorption cycles [22]. Similarly, the efficacy of mass transfer is pivotal as it dictates the extent to which CO_2 molecules engage with the sorbent particles and are subsequently captured.

The aggregate efficiency of an FBR in carbon capture endeavors is contingent upon these transfer processes. By fine-tuning these variables through judicious design and operation, one can achieve heightened capture rates, diminished energy demands, and superior system performance [23]. Such advancements are integral to the advancement of economically viable and environmentally responsible carbon capture technologies.

2.4 Reactor Modeling and Simulation

Modeling and simulation are instrumental in the conceptualization, refinement, and expansion of FBRs for carbon capture applications [24]. Computational Fluid Dynamics (CFD) stands out among computational tools for its capacity to elucidate the intricate phenomena occurring within FBRs [25]. These simulations empower engineers to anticipate the performance of reactors under a diverse array of operating scenarios.

2.4.1 Computational Fluid Dynamics (CFD) Models

CFD models are pivotal for simulating the fluid dynamics, thermal characteristics, and chemical reactions within FBRs. These models employ the Navier–Stokes equations to delineate fluid movement, as well as energy and species conservation equations to project the behavior of fluidized particles [8]. As shown in Fig. 2.4, CFD affords a granular spatial and temporal depiction of flow patterns, thermal profiles, and concentration differentials, which are vital for deciphering and enhancing reactor function.

For the dual-phase nature of FBRs, CFD models typically integrate either an Eulerian-Eulerian or an Eulerian–Lagrangian framework [27]. The Eulerian-Eulerian method regards both fluid and solid phases as continuous but distinct entities, resolving separate momentum equations for each. The Eulerian–Lagrangian method, however, traces discrete particles as they navigate the fluid phase, offering intricate insights at the cost of increased computational intensity [26].

The predictive efficacy of modeling and simulation is indispensable for a myriad of reasons:

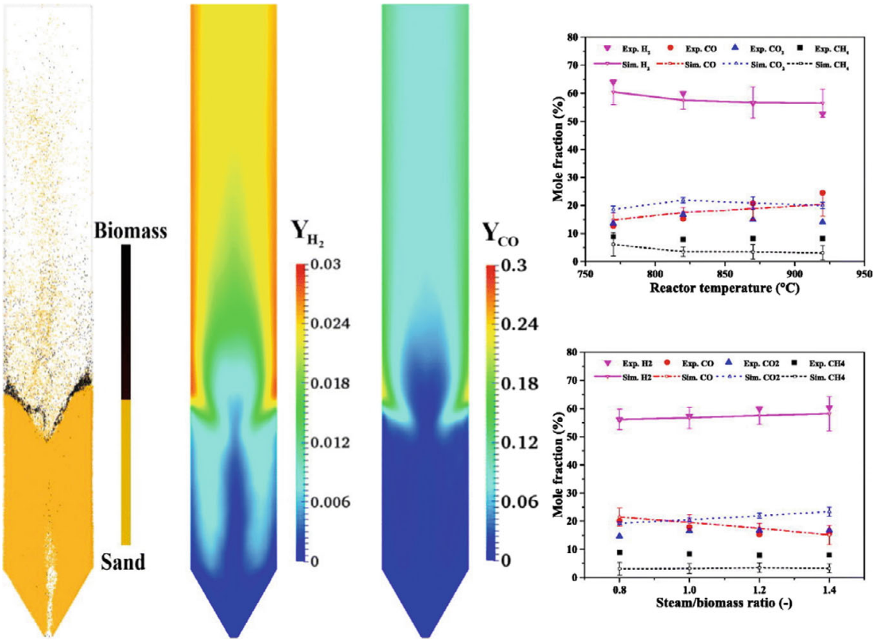


Fig. 2.4 Simulated results for the impact of operating parameters on biomass gasification through the Eulerian–Lagrangian approach (reprinted from ref. [26] with permission of Elsevier)

- Design Parameter Optimization: CFD models enable the virtual exploration of the effects of varying design elements, such as distributor plate configuration or bed elevation, on reactor functionality [26]. This empowers engineers to refine these parameters virtually, obviating the necessity for costly and protracted empirical alterations.
- Fluidization Regime Comprehension: The distinct properties of various fluidization regimes, like bubbling, slugging, or turbulent flows, exert substantial influence on reactor dynamics. CFD models are instrumental in forecasting regime transitions and their consequent effects on reaction rates and homogenization efficacy [28].
- Reaction Kinetics Simulation: These simulations are capable of mimicking the chemical processes transpiring within reactors, including the absorption and rejuvenation cycles of CO_2 on sorbents [29]. By accurately projecting reaction dynamics and conversion efficiencies, CFD assists in enhancing reactor performance for maximal carbon capture.
- Thermal Exchange Evaluation: CFD analyses elucidate the heat transfer attributes of reactors, crucial for processes such as sorbent regeneration [24]. The simulations aid in pinpointing and mitigating thermal inconsistencies.

- Wear and Erosion Projections: FBR components are susceptible to wear and erosion. CFD can forecast zones of intensive particle collisions and attrition, thus informing the design of more durable reactors [30].
- Scale-up Impact Assessment: Laboratory-to-industry scale transitions can significantly alter reactor behavior due to escalated pressure drops, thermal losses, or mixing anomalies [31]. CFD provides essential foresight into these scaling repercussions, facilitating more precise design and operation on an industrial scale.
- Sorbent Behavior Prediction: For carbon capture applications, the performance and reconstitution of sorbents are pivotal. CFD simulations can replicate the thermal and chemical ambiances that sorbents encounter, predicting their reactivity and longevity [32].
- Safety and Regulatory Adherence: CFD models are valuable in preempting the emergence and dispersal of potential thermal anomalies or pollutant concentration zones, thereby contributing to the safe, regulatory-compliant design and operation of FBRs.

2.4.2 Relevance in Scaling Up from Laboratory to Industrial Scale

The amplification from laboratory to industrial scale stands as a formidable challenge in reactor design, wherein modeling and simulation prove to be indispensable tools:

- Foreseeing Scale-up Outcomes: CFD models facilitate projections of the effects that scaling up will have on fluid dynamics and reaction kinetics, thereby illuminating potential deviations in process efficiency observed at smaller scales [31].
- Refinement of Design: Through modeling and simulation, various design frameworks can be virtually tested, conserving time and resources that would otherwise be expended on constructing physical prototypes [33]. This proves especially beneficial in fine-tuning reactor designs for mass production and industrial deployment.
- Risk Reduction: Predictive models play a critical role in preempting the performance of FBRs at escalated scales, enabling the identification and alleviation of prospective risks, thereby smoothing the transition to full-scale operations [34].

In conclusion, the employment of reactor modeling and simulation, with a special emphasis on CFD, is essential in the advancement and refinement of FBR utilized for carbon capture. These computational models impart intricate understandings of the multifaceted events transpiring within FBRs, informing design choices, elevating performance, and facilitating the effective magnification of these vital technologies for carbon capture and the safeguarding of our environment.

Numerous case studies serve as testaments to the efficacy of FBRs in the domain of carbon capture, notably within the power generation sector [35–40]. These examples illustrate that, despite the considerable operational challenges associated with FBRs, such obstacles can be surmounted through meticulous design, judicious material selection, and strategic operational management [15, 41–43]. The lessons gleaned from empirical evidence and ongoing refinement indicate that FBRs are capable of achieving superior performance in carbon capture tasks, thereby playing a significant role in mitigating greenhouse gas emissions across various industries.

In summation, an exploration into the mechanics of FBR underscores the necessity of a profound comprehension of these principles to foster progress in carbon capture technologies. As global efforts intensify to curtail greenhouse gas emissions, the enhancement of FBR technology emerges as an essential avenue toward realizing this ambition. The knowledge distilled from this discourse provides a groundwork for forthcoming innovations in FBR configuration and functionality, propelling us towards a future that is both more sustainable and environmentally benign.

References

1. Daud WRW (2008) Fluidized bed dryers-recent advances. Adv Powder Technol 19:403–418
2. Li Z, Li J, Liu X, Chen R (2021) Progress in enhanced fluidization process for particle coating via atomic layer deposition. Chem Eng Process-Process Intensif 159:108234
3. Mörl L, Heinrich S, Peglow M (2007) Fluidized bed spray granulation. Elsevier
4. Chaudhury S, Apelian D (2006) Fluidized bed heat treatment of cast Al-Si-Cu-Mg alloys. Metall and Mater Trans A 37:2295–2311
5. Xue Y, Zhou S, Brown RC, Kelkar A, Bai X (2015) Fast pyrolysis of biomass and waste plastic in a fluidized bed reactor. Fuel 156:40–46
6. Tezer Ö, Karabağ N, Öngen A, Çolpan CÖ, Ayol A (2022) Biomass gasification for sustainable energy production: a review. Int J Hydrogen Energy 47:15419–15433
7. Ma Z, Glatzmaier G, Mehos M (2014) Fluidized bed technology for concentrating solar power with thermal energy storage. J Sol Energy Eng 136:031014
8. Gómez-Barea A, Leckner B (2010) Modeling of biomass gasification in fluidized bed. Prog Energy Combust Sci 36:444–509
9. Geldart D (1973) Types of gas fluidization. Powder Technol 7:285–292
10. Cocco R, Chew JW (2023) 50 years of Geldart classification. Powder Technol 428:118861
11. Kwauk M, Li J (1996) Fluidization regimes. Powder Technol 87:193–202
12. Leckner B (1998) Fluidized bed combustion: mixing and pollutant limitation. Prog Energy Combust Sci 24:31–61
13. Kim K, Park Y-K, Park J, Jung E, Seo H, Kim H et al (2014) Performance comparison of moving and fluidized bed sorption systems for an energy-efficient solid sorbent-based carbon capture process. Energy Procedia 63:1151–1161
14. Oka S (2003) Fluidized bed combustion. CRC Press
15. Seddighi S, Clough PT, Anthony EJ, Hughes RW, Lu P (2018) Scale-up challenges and opportunities for carbon capture by oxy-fuel circulating fluidized beds. Appl Energy 232:527–542
16. Kim SD, Kang Y (1997) Heat and mass transfer in three-phase fluidized-bed reactors-an overview. Chem Eng Sci 52:3639–3660
17. Chen JC, Grace JR, Golriz MR (2005) Heat transfer in fluidized beds: design methods. Powder Technol 150:123–132
18. Stenberg V, Sköldberg V, Öhrby L, Rydén M (2019) Evaluation of bed-to-tube surface heat transfer coefficient for a horizontal tube in bubbling fluidized bed at high temperature. Powder Technol 352:488–500

19. Hou B, Tang H, Zhang H, Shao G, Li H, Zhu Q (2013) Experimental and theoretical investigation of mass transfer in a circulating fluidized bed. Chem Eng Sci 102:354–364
20. Gómez-Barea A, Ollero P, Leckner B (2007) Mass transport effects during measurements of gas–solid reaction kinetics in a fluidised bed. Chem Eng Sci 62:1477–1493
21. Dieter H, Bidwe AR, Varela-Duelli G, Charitos A, Hawthorne C, Scheffknecht G (2014) Development of the calcium looping CO_2 capture technology from lab to pilot scale at IFK, University of Stuttgart. Fuel 127:23–37
22. McDonough JR, Law R, Reay DA, Zivkovic V (2018) Intensified carbon capture using adsorption: heat transfer challenges and potential solutions. Therm Sci Eng Prog 8:17–30
23. Dhoke C, Zaabout A, Cloete S, Seo H, Park Y-k, Demoulin L et al (2020) Demonstration of the novel swing adsorption reactor cluster concept in a multistage fluidized bed with heat-transfer surfaces for postcombustion CO_2 capture. Ind Eng Chem Res 59:22281–22291
24. Ju Y, Lee C-H (2019) Dynamic modeling of a dual fluidized-bed system with the circulation of dry sorbent for CO_2 capture. Appl Energy 241:640–651
25. Chalermsinsuwan B, Piumsomboon P, Gidaspow D (2010) A computational fluid dynamics design of a carbon dioxide sorption circulating fluidized bed. AIChE J 56:2805–2824
26. Wang S, Luo K, Hu C, Sun L, Fan J (2018) Impact of operating parameters on biomass gasification in a fluidized bed reactor: an eulerian-lagrangian approach. Powder Technol 333:304–316
27. Chiesa M, Mathiesen V, Melheim JA, Halvorsen B (2005) Numerical simulation of particulate flow by the Eulerian-Lagrangian and the Eulerian-Eulerian approach with application to a fluidized bed. Comput Chem Eng 29:291–304
28. Xie N, Battaglia F, Pannala S (2008) Effects of using two-versus three-dimensional computational modeling of fluidized beds-part I, hydrodynamics. Powder Technol 182:1–13
29. Zimmermann S, Taghipour F (2005) CFD modeling of the hydrodynamics and reaction kinetics of FCC fluidized-bed reactors. Ind Eng Chem Res 44:9818–9827
30. Blaser PJ, Corina G (2012) Validation and application of computational modeling to reduce erosion in a circulating fluidized bed boiler. Int J Chem Reactor Eng 10
31. Werther J (1992) Scale-up modeling for fluidized bed reactors. Chem Eng Sci 47:2457–2462
32. Sattari F, Tahmasebpoor M, Valverde JM, Ortiz C, Mohammadpourfard M (2021) Modelling of a fluidized bed carbonator reactor for post-combustion CO_2 capture considering bed hydrodynamics and sorbent characteristics. Chem Eng J 406:126762
33. Singh RI, Brink A, Hupa M (2013) CFD modeling to study fluidized bed combustion and gasification. Appl Therm Eng 52:585–614
34. Matsen JM (1996) Scale-up of fluidized bed processes: principle and practice. Powder Technol 88:237–244
35. Hawthorne C, Dieter H, Bidwe A, Schuster A, Scheffknecht G, Unterberger S et al (2011) CO_2 capture with CaO in a 200 kWth dual fluidized bed pilot plant. Energy Procedia 4:441–448
36. Veneman R, Li Z, Hogendoorn J, Kersten SR, Brilman DWF (2012) Continuous CO_2 capture in a circulating fluidized bed using supported amine sorbents. Chem Eng J 207:18–26
37. Sjostrom S, Krutka H, Starns T, Campbell T (2011) Pilot test results of post-combustion CO_2 capture using solid sorbents. Energy Procedia 4:1584–1592
38. Li H, Li S, Ren Q, Li W, Xu M, Zhang Liu J et al (2014) Experimental results for oxy-fuel combustion with high oxygen concentration in a 1 MWth pilot-scale circulating fluidized bed. Energy Procedia 63:362–371
39. Ströhle J, Orth M, Epple B (2014) Design and operation of a 1 MWth chemical looping plant. Appl Energy 113:1490–1495
40. Damen K, Gnutek R, Kaptein J, Nannan NR, Oyarzun B, Trapp C et al (2011) Developments in the pre-combustion CO_2 capture pilot plant at the Buggenum IGCC. Energy Procedia 4:1214–1221
41. Xu D, Tong A, Fan L-S (2022) State of scale-up development in chemical looping technology for biomass conversions: a review and perspectives. Waste Biomass Valorization 13:1363–1383
42. Ylätalo J, Ritvanen J, Tynjälä T, Hyppänen T (2014) Model based scale-up study of the calcium looping process. Fuel 115:329–337

43. Otitoju O, Oko E, Wang M (2020) A new method for scale-up of solvent-based post-combustion carbon capture process with packed columns. Int J Greenhouse Gas Control 93:102900

Open Access This chapter is licensed under the terms of the Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/), which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license and indicate if changes were made.

The images or other third party material in this chapter are included in the chapter's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the chapter's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.

Chapter 3
Gasification in FBRs

3.1 Introduction to Fluidized Bed Gasification

Pre-combustion carbon capture, also known as pre-combustion CO_2 capture or gasification, is an emerging technology for CO_2 emission reduction (see Fig. 3.1). The core of this method lies in separating the carbon-containing part from the fuel before combustion through gasification, thereby reducing or avoiding carbon emissions [1]. This approach has significant advantages over traditional post-combustion capture techniques. In traditional post-combustion capture schemes, the mixed gases produced after the complete combustion of fuel require a complex treatment process to separate CO_2, which is both energy-intensive and costly [2]. Gasification completes the separation of carbon before the fuel is burned, which not only reduces the treatment cost but also enhances the energy conversion efficiency [3].

In the context of FBRs, pre-combustion CO_2 capture, or gasification, is typically achieved through a water–gas shift reaction, where carbon monoxide (CO) in the syngas reacts with steam to produce additional hydrogen and CO_2 [4]. The resulting CO_2 can be captured from the gas stream using physical or chemical solvents, allowing for the production of hydrogen-rich syngas, which can be utilized as a versatile energy carrier or as a feedstock for producing various chemicals [5]. High-temperature conditions in fluidized bed gasification (FBG) are conducive to the capture and conversion of CO_2 [6, 7]. Recent studies have demonstrated the feasibility of integrating sorbent-based capture systems directly into the fluidized bed, thereby enhancing the efficiency of CO_2 capture and reducing the energy penalty associated with gas separation [8].

However, despite the potential of FBG technology in CO_2 capture, these technologies are still in the development stage and require further research and demonstration projects to verify their economic and practical viability [9]. For instance, operating pressure and CO_2 concentration during the FBGs have a significant impact on reaction performance, and these factors need to be optimized for practical applications. Key research has focused on optimizing the gasification process to enhance syngas

© The Author(s) 2025
L. Duan and D. Lu, *Fluidized Bed Reactors for Carbon Capture*,
https://doi.org/10.1007/978-981-96-0274-2_3

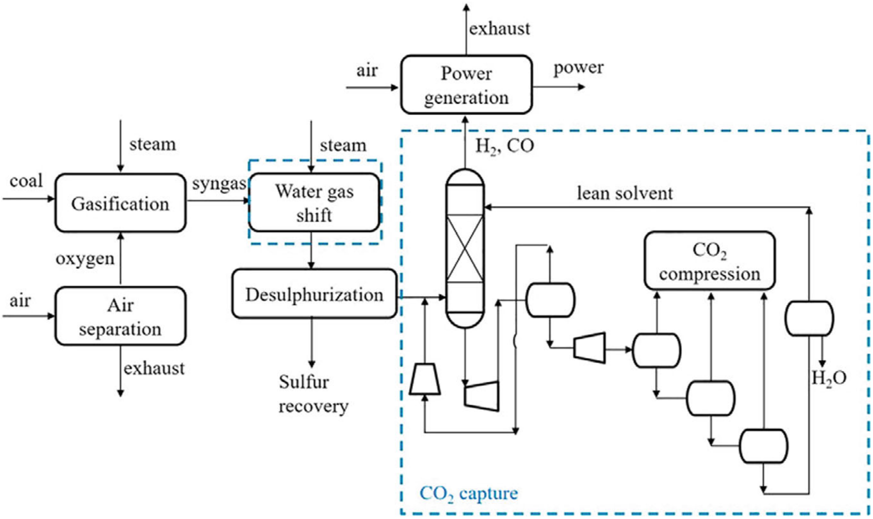


Fig. 3.1 Process flow diagram illustrating the stages of pre-combustion CO_2 capture in a fluidized bed gasifier (from ref. [10], licensed under CC-BY 4.0)

quality, increase carbon conversion efficiency, and integrate effective CO_2 capture mechanisms.

3.2 Advantages of Gasification in FBRs

Gasification in FBR offers several distinct advantages over other types of gasifiers, such as fixed-bed, fluidized-bed, or dual-fluidized bed gasifiers (see Fig. 3.2), particularly in terms of operational flexibility, scalability, and environmental performance. One of the primary benefits is the high efficiency of gasification due to the excellent heat characteristics and mass transfer of the fluidized bed. This results in uniform temperature distribution, which minimizes hot spots and allows for a more complete conversion of the feedstock into syngas [11].

Another principal advantage of FBRs is their ability to handle a variety of feedstocks with different properties such as size, shape, and moisture content. This versatility is crucial for utilizing low-cost, locally available, and low-grade materials, which might otherwise be considered waste. Moreover, the fluidized bed enables uniform temperature distribution throughout the reactor, preventing hot spots that can lead to the formation of undesirable by-products like tar and methane.

Furthermore, a significant advantage of using FBRs in gasification processes is their enhanced reaction kinetics. The intense mixing of gas and solid particles in the fluidized state ensures constant contact between the reactants, leading to faster

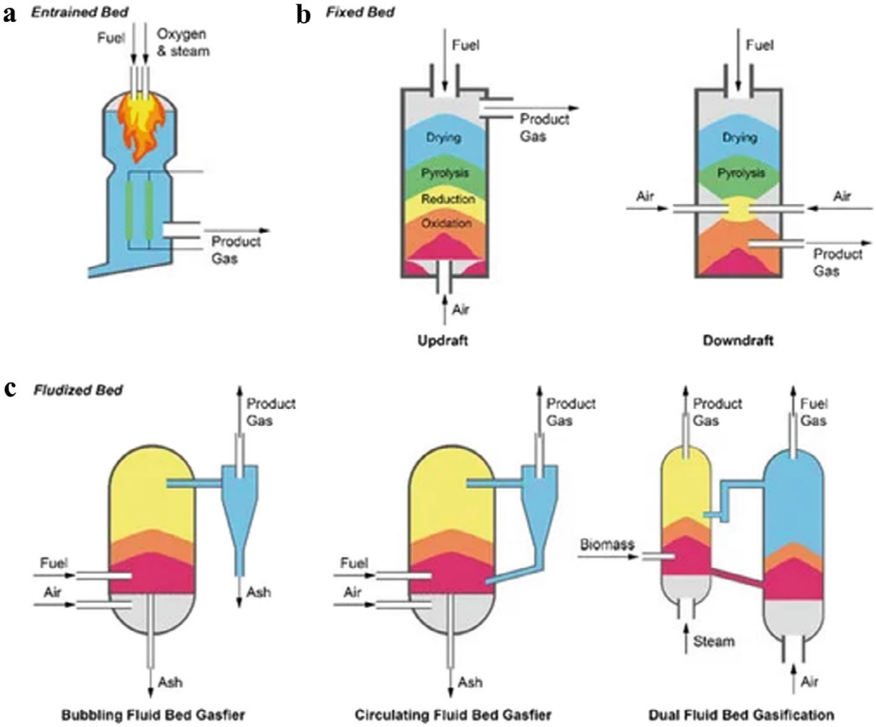


Fig. 3.2 Schematic illustration of different types of gasifiers: **a** entrained flow, **b** fixed bed (left: updraft and right: downdraft) and **c** fluidized bed (left: bubbling fluidized bed, middle: CFB and right: dual fluidized bed) (from ref. [12], licensed under CC-BY 4.0)

reaction rates and higher efficiencies in syngas production. Furthermore, the operational conditions in FBRs, such as temperature, pressure, and residence time, can be precisely controlled to optimize the gasification reactions and improve the quality of the syngas produced (see Fig. 3.3).

Environmental benefits are also notable in FBR gasification. The technology facilitates the capture of CO_2, a critical factor in mitigating climate change. By integrating CO_2 capture and sequestration systems, FBRs can significantly reduce the amount of CO_2 released into the atmosphere from gasification processes [14]. Additionally, the reduction in the emission of other pollutants, such as sulfur oxides (SO_x) and nitrogen oxides (NO_x), can be achieved through careful control of the gasification environment, making FBRs an environmentally friendly option in energy production.

Finally, the scalability of fluidized bed gasifiers is another significant advantage. They are more readily scalable to larger capacities than fixed bed designs, making them suitable for industrial applications and larger energy production scenarios. The economic feasibility of fluidized bed gasifiers is enhanced by their operational efficiencies and the potential for integration with other processes, such as combined heat

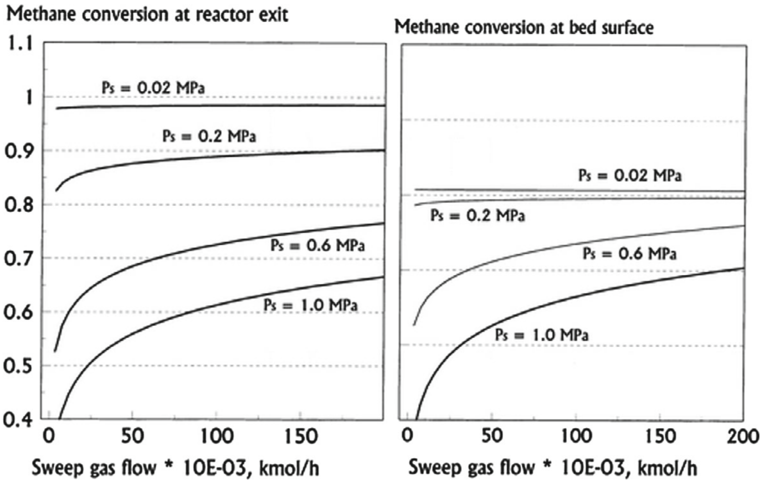


Fig. 3.3 Effect of permeate side pressure and sweep gas flow rate on methane conversion at the bed surface and reactor exit (reprinted from ref. [13] with permission of American Chemical Society)

and power (CHP) systems, which can utilize the heat generated during gasification, further improving the overall energy efficiency of the system.

3.3 Recent Progress of CO_2 Capture During Gasification in FBRs

The latest developments in FBG technology in the field of pre-combustion CO_2 capture mainly include the application of modified adsorbents, the continuous capture capability of multi-stage fluidized bed systems, the study of calcium oxide (CaO) as an adsorbent, simulation research on circulating fluidized bed reactors (CFBRs), and the successful experiment of continuous CO_2 adsorption–desorption demonstration units. These advancements provide significant support for improving CO_2 capture efficiency and promoting the industrial application of the technology. These developments in FBG technology are mainly reflected in the following aspects:

- The application of modified adsorbents has improved the efficiency of CO_2 capture. Experimental measurements have shown that the CO_2 capture capacity of modified adsorbents in a fluidized bed increases significantly with the increase of solid circulation rate, increase of solid hold up as well as contact time (see Fig. 3.4) [15].

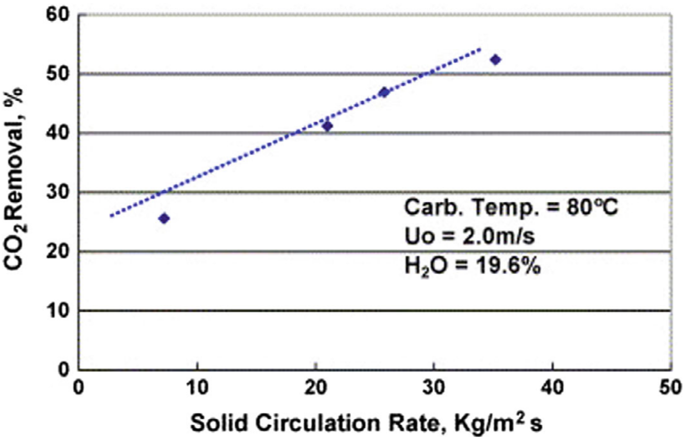


Fig. 3.4 Graph showing the increase in CO_2 capture capacity with solid circulation rate in fluidized beds (reprinted from ref. [15] with permission of Elsevier)

- A multi-stage fluidized bed system can achieve continuous CO_2 capture, and laboratory-scale experiments have already reached the designed capture targets. This indicates that the system can effectively capture CO_2 in a short time under conditions of sufficient gas–solid contact and rapid adsorption kinetics [16].
- In biomass air–steam FBG, CaO as an adsorbent for CO_2 in the synthesis gas has been studied for its feasibility through reaction kinetics modeling methods. This study provides important theoretical support for the development of biomass gasification-CO_2 capture technology (see Fig. 3.5) [17].

- Studies on CFBRs have shown that simulation research can better understand the factors affecting the CO_2 removal rate, such as particle volume fraction, CO_2 mass fraction, and solid temperature distribution characteristics [18].

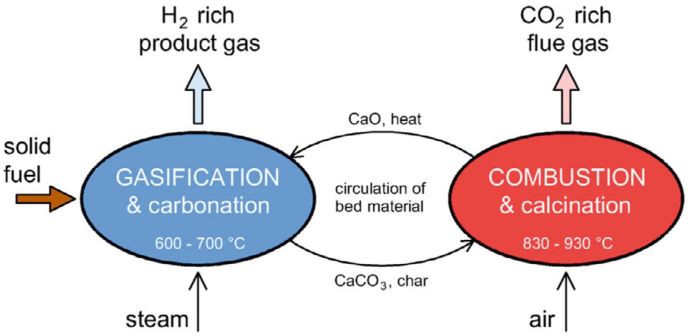


Fig. 3.5 Principle of gasification with in-situ CO_2 capture in biomass gasification [17]

- The role of computational fluid dynamics (CFD) in optimizing gasifier designs cannot be understated [19]. CFD techniques have been instrumental in studying the hydrodynamics and reaction kinetics within fluidized beds, leading to better predictive capabilities and design improvements. These advancements are crucial for scaling up the technology and ensuring consistent performance across different scales and configurations.
- Advancements in technology have also focused on optimizing the variables to improve efficiency and adapt to various feedstocks. For instance, in the pilot studies with different types of wastes, temperatures in the range of 770–870 °C have shown significant variations in gas yield and quality based on the air ratio and feedstock type [20].

Technologies like the fluidized bed two-stage gasification process have demonstrated potential for high efficiency and low tar content in the produced syngas. This process, which combines a pyrolyzer and a gasifier, has shown that maintaining reaction temperatures around 700 °C in the pyrolyzer and 850 °C in the gasifier can significantly enhance the heating value of the produced gas while keeping tar content to a minimum (see Fig. 3.6) [21]. This is crucial for the economic viability of using syngas for power generation or as a chemical feedstock.

Moreover, the incorporation of catalysts into the fluidized bed to enhance CO_2 capture and improve gasification efficiency has been a focus of recent research. Catalysts can help reform tar into syngas and capture CO_2 as stable carbonates,

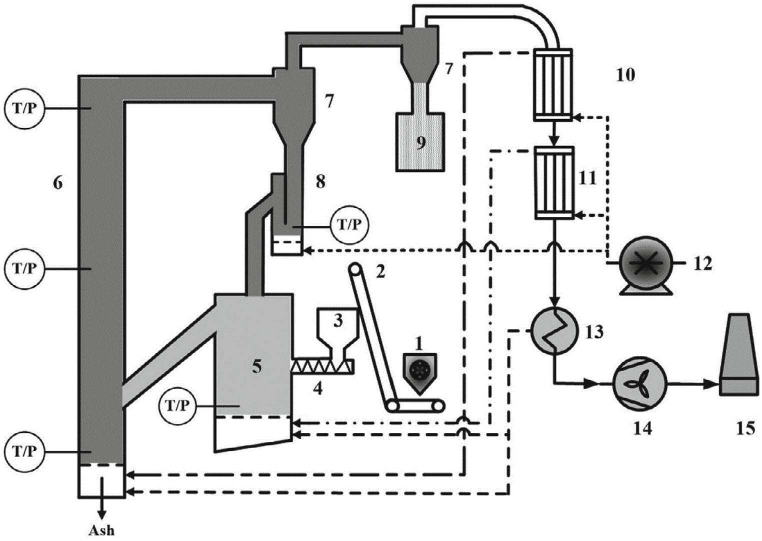


Fig. 3.6 Schematic diagram of the employed experimental dual FBG facility (1-herb residue, 2-conveyor, 3-hopper, 4-screw feeder, 5-pyrolyzer, 6-gasifier, 7-primary cyclone, 8-loop seal, 9-secondary cyclone, 10-primary heat exchanger, 11-secondary heat exchanger, 12-air compressor, 13-boiler, 14 draught fan, 15-chimney) (reprinted from ref. [21] with permission of Elsevier)

which can then be sequestered or used in other processes. The challenge lies in selecting appropriate catalysts that can withstand the harsh conditions inside the gasifier and do not contribute to bed agglomeration or sintering.

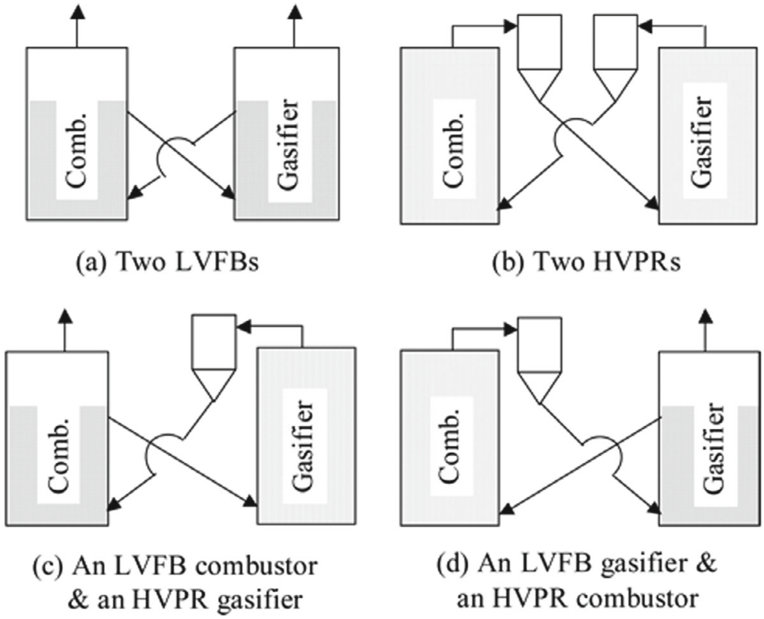
The integration of these CO_2 capture technologies into FBR gasification processes not only helps in reducing CO_2 emissions but also improves the economic viability of these systems by potentially generating tradable carbon credits. Moreover, ongoing research is focusing on developing advanced materials and processes that can further enhance the efficiency and reduce the operational costs of CO_2 capture in FBRs.

3.4 Design and Scale-Up Challenges

Designing and scaling up FBRs for gasification and CO_2 capture presents a set of complex challenges that need careful consideration to ensure operational efficiency and economic viability. These challenges are predominantly related to feedstock variability, bed material lifespan, reactor design, process integration, and economic factors.

- **Feedstock variability**: Feedstock variability can significantly impact the gasification process. Different materials behave differently in the reactor, affecting fluidization, heat transfer, and reaction kinetics. Developing gasifiers that can handle a wide range of feedstocks without requiring extensive adjustments to operating conditions is crucial for flexibility and economic viability [22].
- **Bed material lifespan**: Bed material selection and management are also critical. The material must facilitate good fluidization and heat transfer without degrading quickly. It must also resist agglomeration and fouling, which can reduce efficiency and lead to operational problems. Advances in bed material technology, such as the use of engineered particles or additives that enhance performance and lifespan, are essential for the successful scale-up of fluidized bed gasifiers.
- **Reactor Design**: The design of FBRs must address the fluid dynamics of the particle-gas mixture, ensuring uniform fluidization and minimizing elutriation of fine particles, which can lead to operational inefficiencies and loss of feed material. The reactor should also be designed to allow for easy integration of CO_2 capture technologies, whether they are based on physical absorption, chemical looping, or membrane separation. The scalability of the reactor design from pilot to commercial scale is another critical aspect, requiring a deep understanding of scaling laws on reactor performance [14]. Several different technical options for dual FBG (DFBG) technology are shown in Fig. 3.7.

- **Process Integration**: Integrating the gasification process with CO_2 capture and sequestration systems presents challenges in terms of process optimization and control (see Fig. 3.8). The coupling of these processes must be managed to minimize energy penalties associated with CO_2 capture and to ensure that the overall



(a) Two LVFBs

(b) Two HVPRs

(c) An LVFB combustor & an HVPR gasifier

(d) An LVFB gasifier & an HVPR combustor

Fig. 3.7 Four technical options for dual FBG (DFBG) technology under different combinations of low-velocity fluidized bed (LVFB) and high-velocity pneumatic riser (HVPR) (reprinted from ref. [23] with permission of American Chemical Society)

system remains economically viable. Advanced control strategies, such as real-time process monitoring and adaptive control systems, are essential for optimizing the operation under varying feedstock and operating conditions.

- **Economic Factors**: The economic viability of FBRs for gasification and CO_2 capture depends on various factors, including capital costs, operational costs, feedstock availability, and energy market conditions. The high initial capital investment in advanced materials and sophisticated control systems can be a significant barrier to deployment. However, operational cost savings from increased efficiency, along with potential revenues from by-products and carbon credits, can offset these costs. Economic assessments must consider these factors comprehensively to determine the feasibility of scaling up FBR technologies.

Finally, the mechanical design of large-scale fluidized bed gasifiers must address issues like wear and tear, thermal stresses, and material fatigue. The operational environment inside a gasifier is harsh, with high temperatures, corrosive atmospheres, and abrasive materials. Developing materials and designs that can withstand these conditions over long operational periods is critical to the long-term success and economic viability of these systems.

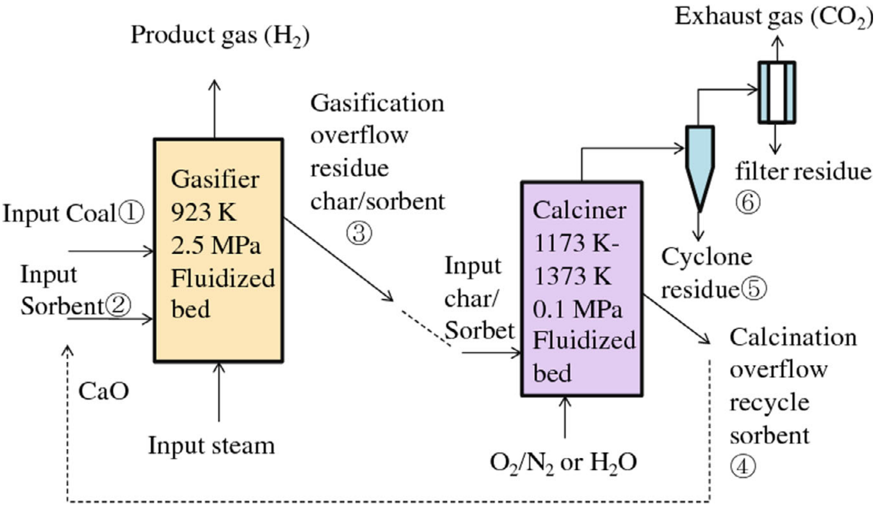


Fig. 3.8 Scheme of integrated gasification system for the in-situ CO_2 capture coal gasification (reprinted from ref. [24] with permission of Elsevier)

3.5 Case Studies and Performance Evaluation

To demonstrate the practical application and effectiveness of FBRs in gasification and CO_2 capture, several case studies can be reviewed. These case studies highlight the operational performance, challenges faced, and the solutions implemented to enhance system efficiency and sustainability.

- **Case study 1: Biomass gasification in a pilot-scale FBR**: A pilot-scale study conducted in Austria focused on the gasification of biomass using an FBR integrated with in situ CO_2 capture technology. The study aimed to evaluate the syngas quality, carbon conversion efficiency, and CO_2 capture rate under different operating conditions. The results showed that optimizing the steam to biomass ratio and reactor temperature could significantly enhance syngas quality and increase carbon conversion efficiency (see Fig. 3.9). CO_2 was captured using a chemical looping method, which proved effective in reducing CO_2 emissions by up to 90% [25]. This case study demonstrated the potential of FBRs in converting biomass to high-quality syngas while capturing CO_2 [26].

- **Case study 2: Coal gasification for power generation**: In China, a commercial-scale FBR was used for the gasification of coal to generate electricity. The reactor was designed to handle high throughputs of coal and was equipped with advanced syngas cleaning and CO_2 capture systems. The performance evaluation showed that the FBR could achieve a high gasification efficiency of approximately 80%, with CO_2 capture rates exceeding 85% [28]. The integration of the gasification process with a combined cycle power plant resulted in overall plant efficiencies

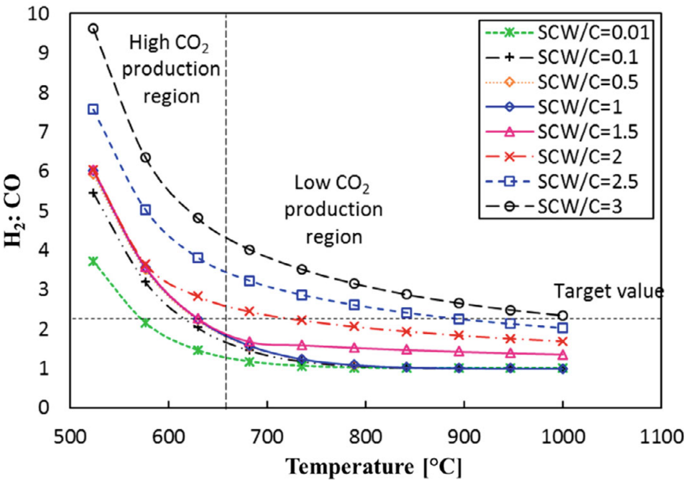


Fig. 3.9 Relationship between fuel characteristic and syngas quality along with CO_2 capture efficiency (from ref. [27], licensed under CC-BY 4.0)

of over 40% (see Fig. 3.10), highlighting the efficiency and scalability of FBR technology in large-scale energy production [28, 29].

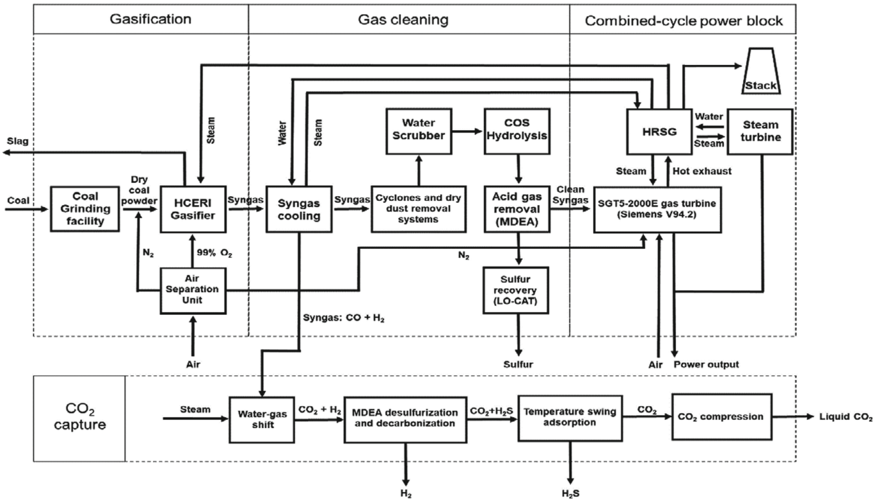


Fig. 3.10 System flow of the GreenGen integrated gasification combined cycle (IGCC) plant with pre-combustion CO_2 capture [28]

- **Case Study 3: Waste-to-energy conversion**: Another case study involved a waste-to-energy plant in Japan where municipal solid waste (MSW) was processed using a fluidized bed gasifier (see Fig. 3.11). The plant utilized an advanced waste preprocessing system to prepare the MSW for gasification. The FBR efficiently converted the waste into syngas, which was then used to generate electricity and heat. CO_2 capture was integrated using a post-combustion capture system, achieving a capture rate of approximately 75%. This case study illustrated the versatility of FBRs in handling diverse feedstocks and their potential to contribute to sustainable waste management and energy production [30].

These case studies provide valuable insights into the operational challenges and technological solutions associated with FBR gasification and CO_2 capture. They underscore the importance of system design, process optimization, and integration of advanced technologies in achieving high efficiencies and environmental benefits.

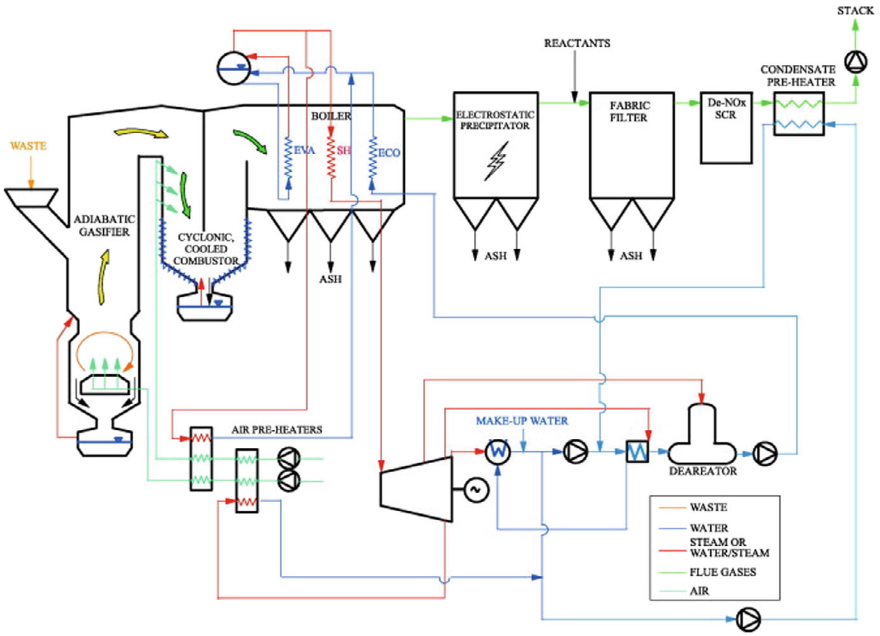


Fig. 3.11 Schematic representation of the low-temperature fluidized-bed gasifier (LTFBG), which was meant to reproduce the two-step oxidation technology proposed by Ebara in Japan [31]

3.6 CO_2 as an Oxidant in FBG

The use of CO_2 as an oxidant in FBG involves the reaction of solid fuels such as biomass or coal with CO_2, facilitated by high temperatures to promote the gasification process of carbon, converting it into gases like carbon monoxide (CO) or other gaseous products. This process not only helps to reduce greenhouse gas emissions but also produces synthesis gas that can be used for energy conversion and chemical production.

The use of CO_2 as a gasification agent in the biomass field has received considerable attention, and while it is largely at the theoretical stage, the studies from Vienna University of Technology have indicated that fluidized bed biomass gasification is effective in converting CO_2 into CO (see Fig. 3.12) [32, 33]. The study experimentally analyzed multiple parameters affecting the CO_2 conversion rate, including temperature and the type of bed material. This suggests that under appropriate operating parameters, FBG can achieve efficient CO_2 conversion.

In certain studies, CO_2 has been partially or completely substituted for steam as a gasification agent to investigate its effects on biomass gasification characteristics (see Fig. 3.13) [34]. The results indicate that substituting CO_2 for steam does not significantly improve gasification efficiency. However, other research has shown that under specific operating conditions and with certain feedstock types, a mixture of $CO_2/H_2O/O_2$ can be used to gasify algal biomass, producing syngas and char [35]. The gasification of lignocellulosic biomass with CO_2 is considered an attractive technology for addressing the rapid depletion of fossil fuels and CO_2 emission reduction [36].

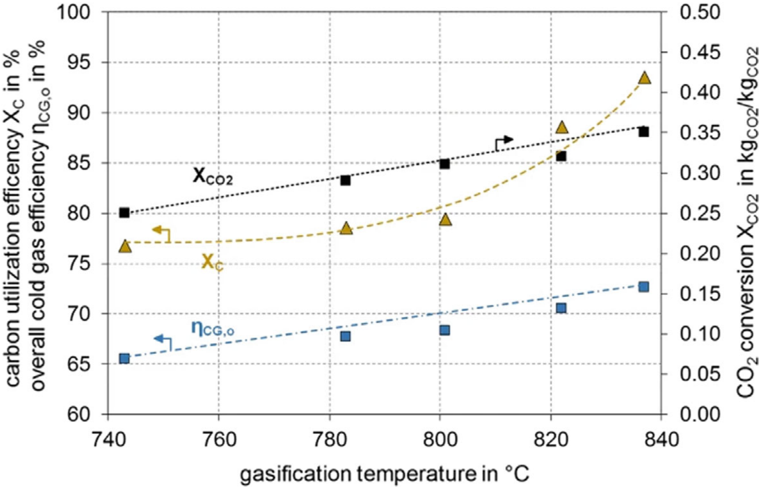


Fig. 3.12 The conversion rate of CO_2 to CO under various operational conditions in a fluidized bed gasifier (from ref. [33], licensed under CC-BY 4.0)

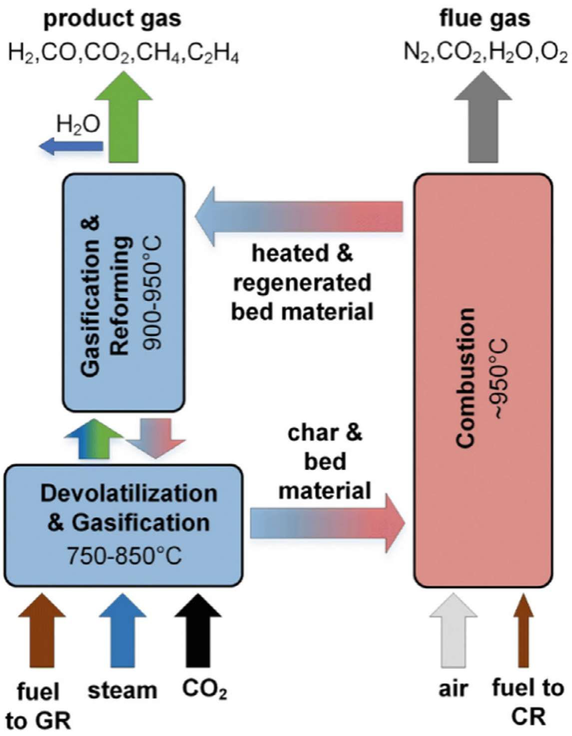


Fig. 3.13 Principle of the advanced dual fluidized bed CO_2/steam gasification system (from ref. [34], licensed under CC-BY 4.0)

The gasification reaction between carbon and CO_2 is a complex multi-step process. For instance, the gasification of char with CO_2 initially involves the formation of carbon–oxygen complexes from CO_2 and char, followed by the desorption of these complexes from char. The microscopic mechanism of this process can be understood in depth through quantum chemical methods.

The role of catalysts in the gasification process is significant. For example, CaO has been found to have a positive catalytic effect on the gasification reaction between carbon and CO_2, influencing the onset temperature, endpoint temperature, and reaction rate (see Fig. 3.14) [37]. This indicates that adding specific catalysts in FBG can optimize the gasification process and improve the efficiency of CO_2 conversion.

Despite the theoretical potential of CO_2 as a gasification agent, practical applications present challenges [39, 40]. For example, one of the challenges in CO_2 electrocatalysis is the selection and design of reactive interfaces that can activate CO_2 to form carbon–carbon bonds with low overpotential. Although this challenge is different from directly using CO_2 as a gasification agent, it reflects the technical difficulties that may be encountered when dealing with CO_2. Another challenge is understanding the molecular mechanisms and key intermediates that control catalytic selectivity [40]. This is crucial for optimizing the application of CO_2 as a gasification agent, as precise control of the reaction process is needed to improve gasification efficiency and product quality.

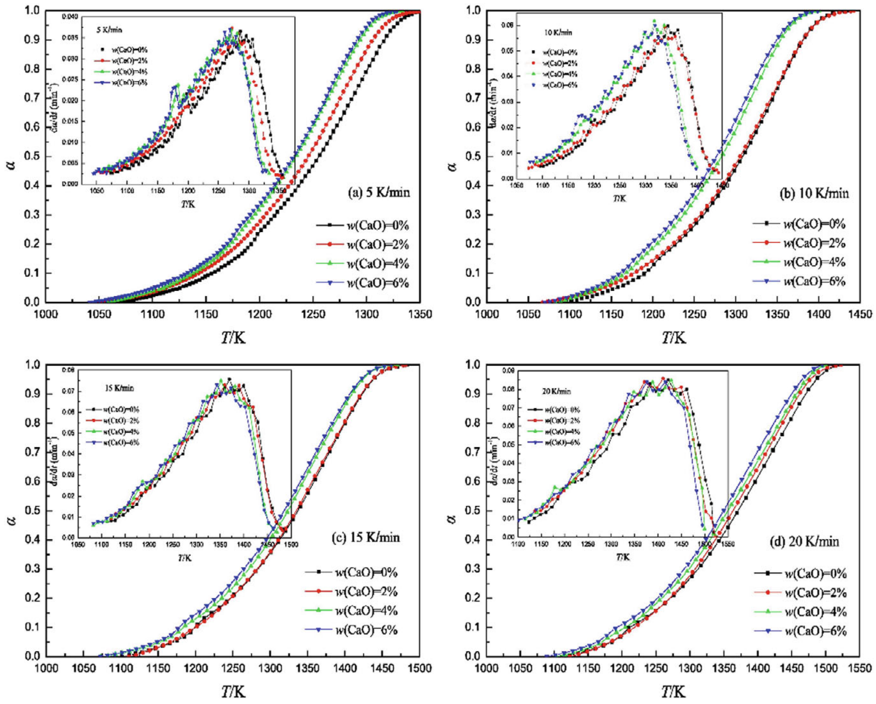


Fig. 3.14 Effects of different CaO ratios on conversion rate curve and conversion rate curve (reprinted from ref. [37, 38] with permission of Elsevier)

In practice, temperature positively affects the gasification reaction, but under certain conditions, such as excessively high temperatures or small particle sizes, it may have adverse effects on gasification outcomes [41]. This means that when using CO_2 as a gasification agent, precise control of operating conditions is necessary to optimize the gasification effect.

The application of CO_2 as an oxidant in FBG demonstrates its potential in the efficient conversion of biomass and coal resources. By precisely controlling reaction conditions and utilizing catalysts, high-efficiency CO_2 conversion can be achieved, providing an effective approach to sustainable energy production and environmental protection.

The environmental impact assessment of FBG technology using CO_2 as an oxidant in practical applications includes the reduction of greenhouse gas emissions, balancing economic and environmental benefits, conducting environmental risk assessments, and implementing life cycle assessments. These studies highlight the potential of the technology in promoting environmental protection and sustainable development in several key areas:

- Reduction of CO_2 Emissions: Projects such as the Shenhua Yulin Coal Deep Processing Base have demonstrated significant CO_2 emission reductions through

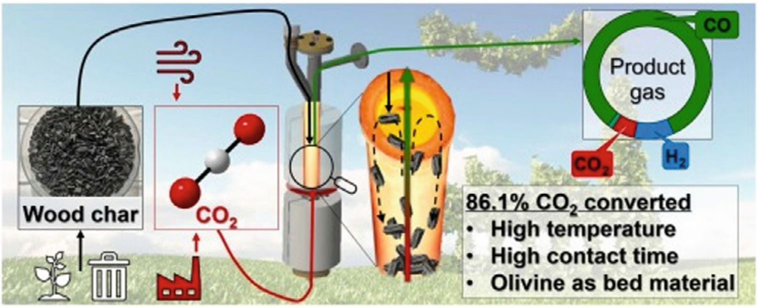


Fig. 3.15 Life cycle assessment of biomass gasification using CO_2 (from ref. [32], licensed under CC-BY 4.0)

energy-saving measures like semi-waste heat recovery processes and the elimination of coal-fired boilers, achieving an annual reduction of 534,300 tons of CO_2 [42]. This indicates that with optimized processes and technological improvements, substantial cuts in greenhouse gas emissions can be made.

- Environmental Risk Assessment: Environmental risk assessments for CCUS projects include steps like defining the scope of the assessment, identifying potential environmental risk sources and receptors [43]. While not directly mentioning FBG, this highlights the importance of systematic environmental risk evaluation, which applies to any technology involving CO_2 handling.
- Life Cycle Assessment (LCA): Studies on the life cycle assessment of power generation systems based on biomass gasification combined cycles provide an LCA of different technological options, examining alternatives in biomass gasification, synthesis gas combustion, and CO_2 emission control [44]. This method helps to understand the overall environmental impact of the technology throughout its life cycle, including GHG emissions (see Fig. 3.15).

It is important to conduct comprehensive assessments that consider not only the direct emissions from the gasification process but also the broader implications of CO_2 utilization and storage, as well as the indirect effects on the economy and society. This holistic approach is crucial for the development and deployment of CO_2-based gasification technologies.

3.7 Future Directions and Research Needs

As the demand for sustainable energy solutions continues to grow, the development of fluidized bed reactors (FBRs) for gasification and CO_2 capture will likely play a critical role in meeting global energy and environmental goals. Looking forward, research and development in FBG will likely focus on several key areas to improve technology and make it more suitable for widespread commercial adoption. These areas include further enhancements to CO_2 capture techniques, the development of more robust and efficient reactor designs, process optimization, and the integration of renewable energy sources to create more comprehensive waste-to-energy or biomass-to-energy solutions.

- **Advancements in CO_2 capture technologies**: Ongoing research is exploring advanced sorbent materials that can capture CO_2 more efficiently and at lower costs. These materials need to be compatible with the high temperatures and corrosive environments found in gasifiers. Additionally, integrating CO_2 capture directly into the gasification process—rather than as a separate post-processing step—could significantly improve the overall efficiency and cost-effectiveness of the technology [14, 45].
- **Reactor design**: Continued innovation in reactor design is also an area of active research, with efforts focusing on improving the reliability and scalability of fluidized bed gasifiers. This includes developing better construction materials that can withstand the harsh conditions inside the reactor, as well as refining the design of components like distributors and cyclones to enhance performance and reduce maintenance requirements [46].
- **Process optimization**: There is a significant need for further optimization of the gasification process to enhance syngas production and carbon conversion rates (see Fig. 3.16). Advanced modeling and simulation tools can provide deeper insights into the complex reactions within FBRs and help in optimizing operational parameters for better performance [12].

- **Integration of renewable energy sources**: Integrating FBR with renewable energy sources, such as solar or wind power, could provide the necessary energy inputs for gasification processes, reducing the reliance on fossil fuels and enhancing the sustainability of the system. These integrated systems could help to balance the intermittent nature of some renewable energy sources and provide a more stable and reliable supply of energy from biomass and waste materials.

3.8 Ending Remarks

The future of FBR technology appears bright, with numerous opportunities for further research and development. As global efforts to combat climate change intensify, the demand for innovative and sustainable energy solutions will undoubtedly increase. FBRs, with their ability to efficiently convert a wide range of feedstocks into clean energy while capturing CO_2, are well-positioned to be at the forefront of these solutions.

FBG, as a pre-combustion CO_2 capture technology, shows promising prospects and development potential. With further technological improvements and scaled-up applications, it is expected to achieve more efficient and cost-effective CO_2 capture and utilization in the future.

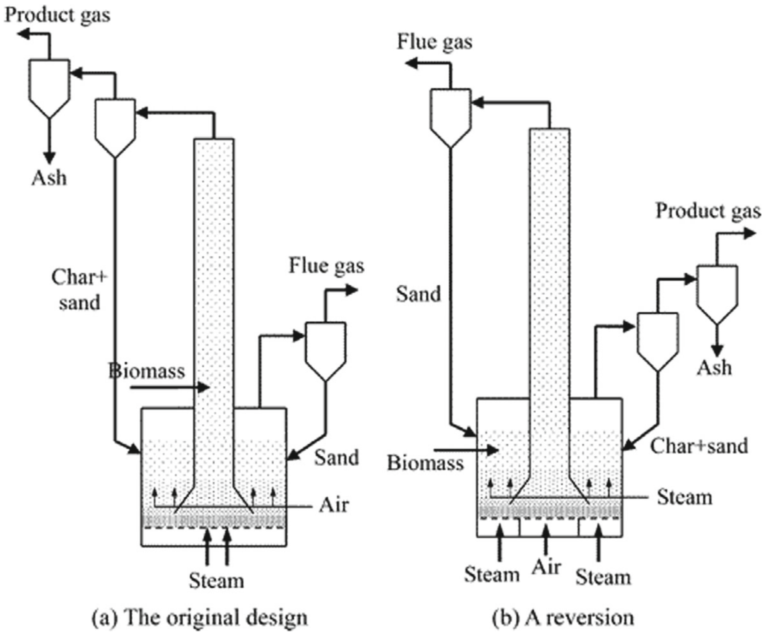


Fig. 3.16 System conception of **a** a recently proposed biomass gasification plant and **b** its reversion (reprinted from ref. [23] with permission of American Chemical Society)

References

1. Corella J, Toledo JM, Molina G (2006) Steam gasification of coal at low−medium (600–800 °C) temperature with simultaneous CO_2 capture in fluidized bed at atmospheric pressure: the effect of inorganic species. 1. Literature review and comments. Ind Eng Chem Res 45:6137–6146
2. Saxena SC, Jotshi CK (1994) Fluidized-bed incineration of waste materials. Prog Energy Combust Sci 20:281–324
3. Miccio M, Miccio F (2013) Conversion of liquid and gaseous fuels in fluidized bed combustion and gasification. Woodhead Publishing
4. Liu Q, Shi Y, Zhong W, Yu A (2019) Co-firing of coal and biomass in oxy-fuel fluidized bed for CO_2 capture: a review of recent advances. Chin J Chem Eng 27:2261–2272
5. Seddighi S, Clough PT, Anthony EJ, Hughes RW, Lu P (2018) Scale-up challenges and opportunities for carbon capture by oxy-fuel circulating fluidized beds. Appl Energy 232:527–542
6. Arena U (2013) Fluidized bed gasification. Woodhead Publishing
7. Materazzi M (2017) Gasification of waste derived fuels in fluidized beds: fundamental aspects and industrial challenges. Springer International Publishing, Cham
8. Kim K, Seo H, Kim DJ, Lee C, Min DY, Kim HM et al (2020) Experimental evaluation of CO_2 capture with an amine impregnated sorbent in dual circulating fluidized bed process. Int J Greenhouse Gas Control 101:103141
9. Theo WL, Lim JS, Hashim H, Mustaffa AA, Ho WS (2016) Review of pre-combustion capture and ionic liquid in carbon capture and storage. Appl Energy 183:1633–1663
10. Xin K, Hashish M, Roghair I, van Sint Annaland M (2020) Process simulation and economic analysis of pre-combustion CO_2 capture with deep eutectic solvents. Front Energy Res 8
11. Antil S, Sachdeva G, Sharma AK (2023) Advancements and challenges in the fluidized bed gasification system: a comperehensive review. J Therm Eng 9

12. Lian Z, Wang Y, Zhang X, Yusuf A, Famiyeh L, Murindababisha D et al (2021) Hydrogen production by fluidized bed reactors: a quantitative perspective using the supervised machine learning approach. J 4:266–287
13. Adris AEM, Grace JR (1997) Characteristics of fluidized-bed membrane reactors: scale-up and practical issues. Ind Eng Chem Res 36:4549–4556
14. Shrestha S, Ali BS, Hamid MDB (2016) Cold flow model of dual fluidized bed: a review. Renew Sustain Energy Rev 53:1529–1548
15. Yi C-K, Jo S-H, Seo Y, Lee J-B, Ryu C-K (2007) Continuous operation of the potassium-based dry sorbent CO_2 capture process with two fluidized-bed reactors. Int J Greenhouse Gas Control 1:31–36
16. Schoeny G, Dietrich F, Fuchs J, Proell T, Hofbauer H (2017) A multi-stage fluidized bed system for continuous CO_2 capture by means of temperature swing adsorption—first results from bench scale experiments. Powder Technol 316:519–527
17. Fuchs J, Müller S, Schmid JC, Hofbauer H (2019) A kinetic model of carbonation and calcination of limestone for sorption enhanced reforming of biomass. Int J Greenhouse Gas Control 90:102787
18. Lu S, Lan B, Xu J, Zhao B, Zou Z, Wang J et al (2023) Optimization of multiple-chamber fluidized beds using coarse-grained CFD-DEM simulations: regulation of solids back-mixing. Powder Technol 428:118886
19. Singh RI, Brink A, Hupa M (2013) CFD modeling to study fluidized bed combustion and gasification. Appl Therm Eng 52:585–614
20. Campoy M, Gómez-Barea A, Ollero P, Nilsson S (2014) Gasification of wastes in a pilot fluidized bed gasifier. Fuel Process Technol 121:63–69
21. Zeng X, Shao R, Wang F, Dong P, Yu J, Xu G (2016) Industrial demonstration plant for the gasification of herb residue by fluidized bed two-stage process. Biores Technol 206:93–98
22. Rüdisüli M, Schildhauer TJ, Biollaz SM, Van Ommen JR (2012) Scale-up of bubbling fluidized bed reactors—a review. Powder Technol 217:21–38
23. Xu GW, Murakami T, Suda T, Matsuzawa Y, Tani H (2006) The superior technical choice for dual fluidized bed gasification. Ind Eng Chem Res 45:2281–2286
24. Lin S, Kiga T, Nakayama K, Suzukib Y (2011) Coal power generation with in-situ CO_2 capture–HyPr-RING method—effect of ash separation on plant efficiency. Energy Procedia 4:378–384
25. Pröll T, Schöny G, Sprachmann G, Hofbauer H (2016) Introduction and evaluation of a double loop staged fluidized bed system for post-combustion CO_2 capture using solid sorbents in a continuous temperature swing adsorption process. Chem Eng Sci 141:166–174
26. Pfeifer C, Puchner B, Hofbauer H (2007) In-situ CO_2-absorption in a dual fluidized bed biomass steam gasifier to produce a hydrogen rich syngas. Int J Chem React Eng 5(1)
27. Sarafraz M, Safaei MR, Jafarian M, Goodarzi M, Arjomandi M (2019) High quality syngas production with supercritical biomass gasification integrated with a water–gas shift reactor. Energies 12:2591
28. Xia C, Ye B, Jiang J, Shu Y (2020) Prospect of near-zero-emission IGCC power plants to decarbonize coal-fired power generation in China: implications from the GreenGen project. J Clean Prod 271:122615
29. Liu H, Ni W, Li Z, Ma L (2008) Strategic thinking on IGCC development in China. Energy Policy 36:1–11
30. Neubacher FP (2013) Waste-to-energy: fluidized bed technology. Springer, New York
31. Consonni S, Viganò F (2012) Waste gasification versus conventional waste-to-energy: a comparative evaluation of two commercial technologies. Waste Manage 32:653–666
32. Müller FJ, Fuchs J, Cuesta MF, Gutiérrez AO, Pratschner S, Müller S et al (2024) CO_2 conversion to CO by fluidized bed biomass gasification: analysis of operational parameters. J of CO_2 Utilization 81:102706
33. Mauerhofer AM, Müller S, Bartik A, Benedikt F, Fuchs J, Hammerschmid M et al (2021) Conversion of CO_2 during the DFB biomass gasification process. Biomass Convers Biorefinery 11:15–27

34. Mauerhofer AM, Mueller S, Benedikt F, Fuchs J, Bartik A, Hofbauer H (2021) CO_2 gasification of biogenic fuels in a dual fluidized bed reactor system. Biomass Convers Biorefinery 11:1101–1116
35. Ebadi AG, Hisoriev H (2019) Gasification of algal biomass (Cladophora glomerata L.) with $CO_2/H_2O/O_2$ in a circulating fluidized bed. Environ Technol 40:749–755
36. Shen Y, Li X, Yao Z, Cui X, Wang C-H (2019) CO_2 gasification of woody biomass: experimental study from a lab-scale reactor to a small-scale autothermal gasifier. Energy 170:497–506
37. Lan C, Liu R, Zhang S, Lyu Q, Gao Y, Yan G (2022) Influence mechanism of CaO on gasification characteristics and kinetic behaviors of carbon-CO_2 reaction. Fuel 311:122583
38. Hervy M, Maistrello J, Brito L, Rizand M, Basset E, Kara Y et al (2021) Power-to-gas: CO_2 methanation in a catalytic fluidized bed reactor at demonstration scale, experimental results and simulation. J CO_2 Utilization 50:101610
39. Kim D, Kley CS, Li Y, Yang P (2017) Copper nanoparticle ensembles for selective electroreduction of CO_2 to C_2–C_3 products. Proc Natl Acad Sci 114:10560–10565
40. Li Y, Kim D, Louisia S, Xie C, Kong Q, Yu S et al (2020) Electrochemically scrambled nanocrystals are catalytically active for CO_2-to-multicarbons. Proc Natl Acad Sci 117:9194–9201
41. Kong D, Wang S, Luo K, Fan J (2023) Numerical study of biomass gasification combined with CO_2 absorption in a bubbling fluidized bed. AIChE J 69:e18096
42. Xiang S (2023) Typical case of greenhouse gas emission environmental impact assessment ⑩ Yulin Coal Deep Processing Base Project. Department of Environmental Impact Assessment and Emission Management, Ministry of Ecology and Environment
43. Goel A, Moghaddam EM, Liu W, He C, Konttinen J (2022) Biomass chemical looping gasification for high-quality syngas: a critical review and technological outlooks. Energy Convers Manage 268:116020
44. Zang G, Zhang J, Jia J, Lora ES, Ratner A (2020) Life cycle assessment of power-generation systems based on biomass integrated gasification combined cycles. Renew Energy 149:336–346
45. Perejón A, Romeo LM, Lara Y, Lisbona P, Martínez A, Valverde JM (2016) The calcium-looping technology for CO_2 capture: on the important roles of energy integration and sorbent behavior. Appl Energy 162:787–807
46. Hanchate N, Ramani S, Mathpati C, Dalvi VH (2021) Biomass gasification using dual fluidized bed gasification systems: a review. J Clean Prod 280:123148

Open Access This chapter is licensed under the terms of the Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/), which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license and indicate if changes were made.

The images or other third party material in this chapter are included in the chapter's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the chapter's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.

Chapter 4
Oxy-Fuel Combustion in FBRs

4.1 Introduction to Oxy-Fuel Combustion

Oxy-fuel combustion, also known as O_2/CO_2 combustion or oxy-fuel, is one of the most promising CO_2 capture technologies for coal-fired power plants, technically or economically. Oxy-fuel combustion represents a pivotal advancement in carbon capture technologies, characterized by fuel burning in an oxygen-rich environment rather than in the air. This process is fundamentally distinct from conventional combustion methods, as it predominantly uses pure oxygen mixed with a recirculated flue gas, predominantly composed of carbon dioxide and water vapor. The primary advantage of this method is its ability to produce a highly concentrated CO_2 stream in the exhaust, significantly simplifying the carbon capture process. By eliminating the dilution effects of nitrogen and other atmospheric gases typically present in air-fired combustion, oxy-fuel combustion enhances the efficiency and efficacy of carbon capture.

Figure 4.1 shows the schematic diagram of a typical oxy-fuel combustion system. During the oxy-fuel process, a mixture of O_2 obtained by the air separation unit (ASU) and part of the recycled flue gases (RFG, predominantly composed of CO_2 and water vapor) is burned with the fuel, thereby increasing the concentration of CO_2 in the exhaust flue gas. So, the principle for oxy-fuel combustion is the substitution of N_2 in air by a gas, usually the RFG.

In the context of FBRs, the implementation of oxy-fuel combustion presents unique advantages. FBRs, known for their excellent heat transfer characteristics and uniform temperature distribution, become even more efficient when combined with oxy-fuel combustion. This synergy results in enhanced combustion efficiency, reduced pollutant emissions, and improved carbon capture rates. The adaptability of FBRs to different fuel types, including low-grade and alternative fuels, further augments their suitability for oxy-fuel combustion processes. This adaptability is particularly crucial in addressing the varying fuel compositions and impurity levels encountered in industrial applications.

© The Author(s) 2025

L. Duan and D. Lu, *Fluidized Bed Reactors for Carbon Capture*,
https://doi.org/10.1007/978-981-96-0274-2_4

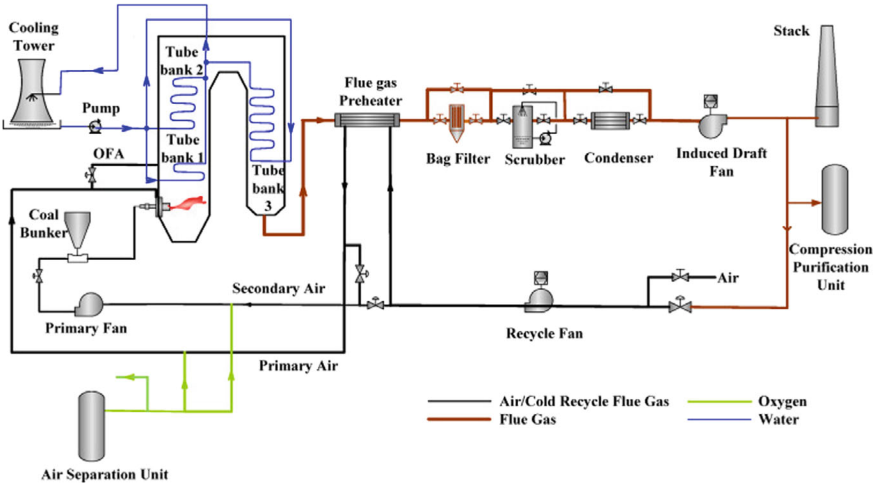


Fig. 4.1 Schematic diagram of the oxy-fuel combustion system [1]

The integration of oxy-fuel combustion in FBRs also necessitates consideration of certain operational aspects, such as the design modifications required to handle the high-temperature environment created by the oxygen-rich atmosphere. Additionally, the recirculation of flue gases in FBR demands precise control systems to maintain optimal combustion conditions and ensure efficient carbon capture. These considerations are critical in the design and operation of FBR utilizing oxy-fuel combustion, highlighting the need for ongoing research and development in this area.

4.2 Technological Developments

The process of using oxygen instead of air to organize combustion with fuel in fluidized beds has a long research history. Fritz Winkler had already used this technology to provide heat for gasifiers almost 100 years ago [2]. Figure 4.2 shows the development history and milestones of the oxy-fuel fluidized bed technology. Horn et al. [3] first proposed the concept of oxy-fuel combustion in the fluidized bed, which was originally used to produce a high-purity CO_2 stream to enhance oil recovery. In the 1980s and 1990s, several laboratory studies were carried out to research the fundamental combustion and pollutant conversion characteristics in an O_2/CO_2 atmosphere. Delft University of Technology [4] and Hokkaido National Industrial Research Institute (HNIRI) [5] conducted oxy-fuel experiments with RFG in the pressurized fluidized bed and atmospheric-pressure bubbling bed. Their results showed that the dry-based CO_2 concentration in flue gas can exceed 90%, verifying the feasibility of the oxy-fuel fluidized bed combustion process. In the past two decades, this technology has achieved rapid development, obtaining a series of patents, as well as several larger capacity demonstration power plants.

One of the most significant benefits of this oxygen-driven process is the dramatic increase in CO_2 concentration in the exhaust gases. In conventional air-fired combustion, CO_2 concentrations typically range from 3–15%, depending on the fuel type and combustion conditions. However, in oxy-fuel combustion, the CO_2 concentration can exceed 90%, greatly facilitating the subsequent carbon capture process. The high CO_2 concentration not only improves the efficiency of post-combustion capture technologies but also reduces the energy penalty associated with carbon capture. This is because the compression-cooling-separation method can be used to purify an extremely large amount of flue gas and obtain liquid CO_2 at a low cost.

In the realm of FBRs, innovations have been particularly focused on optimizing the reactor design to handle the intense heat and corrosive environment created by oxy-fuel combustion. This includes the development of advanced materials and coatings that can withstand high temperatures and prevent corrosion, thereby extending the lifespan of the reactor components [6, 7]. Additionally, there has been significant progress in the control systems for FBRs [8, 9], which are now more capable of maintaining stable and efficient combustion conditions even with the variable fuel qualities and flow rates.

In conclusion, the technological developments in oxy-fuel combustion, particularly when applied to FBRs, represent a significant stride towards more efficient and sustainable carbon capture solutions. These advancements not only enhance the performance of FBRs in carbon capture applications but also contribute to the broader goal of reducing greenhouse gas emissions in energy and industrial sectors.

4.3 Performance of the Oxy-Fuel Fluidized Bed

Compared with conventional air mode, the fuel burned in O_2/RFG instead of O_2/N_2 in an oxy-fuel fluidized bed, and the atmosphere greatly changes fuel combustion characteristics, heat transfer, and the conversion path of pollutants. CO_2 has a larger specific heat capacity than N_2, and the diffusion coefficient of O_2 in CO_2 is smaller than that in N_2 [10]. The change of atmosphere has a significant impact on the combustion of fuel particles in fluidized beds. Take coal particles as an example, the volatile matter release rate in 21% O_2/79% CO_2 atmosphere decreases slightly than in air, while the ignition delay time increases. Also, the peak combustion temperature decreases in 21% O_2/79% CO_2 atmosphere, and it takes a longer time to reach the peak temperature [11, 12]. In addition, the high concentration of CO_2 will cause a gasification reaction with coal char in the oxy-combustion process. Even though the gasification rate is slower than that of the oxidation reaction, the influence of gasification at low O_2 fraction and high bed temperature still cannot be ignored [13, 14].

Table 4.1 briefs the pollutant emissions from oxy-CFB combustion. The experimental results show that the total emission mass of NO_x in oxy-fuel CFB is much

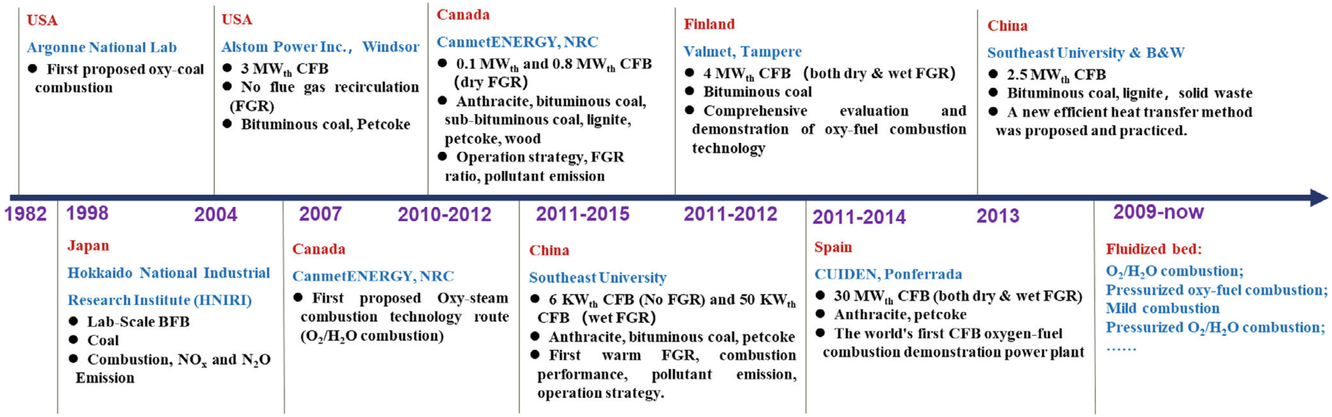


Fig. 4.2 Milestones of the oxy-fuel fluidized bed technology

lower than that in air-combustion mode [15–17]. This is because flue gas recycling greatly extends the residence time of NO_x in the reducing zone [18, 19], and enhances the reduction reactions of NO_x. For SO_2 emission, the formation of SO_2 is thermodynamically favored in high temperature and high inlet oxygen concentration conditions. Also, the flue gas recycling accumulates SO_2 concentration in the flue gas. Limestone is widely used in the in-furnace desulfurization of CFB boilers, and the desulfurization process of limestone includes direct desulfurization and indirect desulfurization depending on the operating temperature and the partial pressure of CO_2, which is different from that in air-fired condition. With high partial pressure of CO_2, the limestone can react directly with SO_2, which is called direct desulfurization. The efficiency of direct desulfurization is revealed to be lower than indirect desulfurization due to the dense pores and low porosity of $CaCO_3$ [20].

4.4 Heat Transfer and Load Distribution

The heat transfer characteristics are the fundamental basis for boiler design. Convective heat transfer of particles and gas is dominant in the dense zone of the fluidized bed, and external heat exchangers (EHEs) can be the suitable choice to increase the heating surface and ensure the stability of the furnace temperature [30, 31]. The immersed heating tubes are adopted in EHEs, and their heat transfer coefficient is larger than other heating surfaces [18, 32, 33].

In the oxy-combustion mode, the share of tri-atomic gases with a high emission rate in the flue gas is higher than in the air mode, so the radiative heat transfer of the flue gas increases at the same temperature. This difference is particularly apparent for oxy-fuel pulverized coal furnaces with high combustion and flue gas temperatures. However, for fluidized bed boilers, whether in the dilute phase region or the flue duct, the enhancing effect of three-atom radiation on heat transfer is not significant due to the lower flue gas temperature [18, 34].

Heat transfer in the dilute phase region is mainly dominated by gas convection and particle radiation, while for flue ducts it is mainly carried out through gas convection. Due to the increased proportion of tri-atomic gases in the oxy-fuel flue gas, the change of physical parameters enhances the convective heat transfer performance, and the heat transfer coefficient of each convective heating surface is higher than that of the air-combustion mode [35, 36].

A dominant parameter of oxy-combustion fluidized bed is the inlet O_2 concentration of the boiler, which directly relates to heat transfer characteristics, arrangement of heating surfaces, and further influences boiler design. The oxy-fuel process can be adapted to both newly designed and retrofit boilers, but in these two cases, there will be significant differences in the selection of oxygen concentration. In the retrofit case, it is necessary to minimize the modification of the original boiler structure, especially for the heating surface. So, the key issue is to comprehensively consider the impact of combustion mode on heat distribution, furnace temperature, and heat transfer characteristics. An oxygen concentration of 27 to 30% is usually selected for

Table 4.1 Pollutant emissions from oxy-CFB combustion with RFG—a summary

Authors	Test facility	Fuels	Combustion conditions	Remarks
Jia et al. [21, 22]	0.1 and 0.8 MWth CFB	Anthracite, bituminous coal, petroleum coke	Air, 28~45% O_2/RFG	• NO_x emissions were lower than in air-firing mode even at temperatures as high as 950 °C in the fluidized bed • CO concentration was the same for air firing and oxyfuel firing with flue gas recirculation under CFBC conditions
Duan et al. [19]	50 kWth CFB	Bituminous coal, petroleum coke	Air, 21% O_2/RFG	• The Ca utilization rate is higher in oxy-fuel combustion than in air combustion, and NO emission is much lower in oxy-fuel combustion
de las Obras-Loscertales et al. [23]	3 kWth BFB	Anthracite coal	35% O_2/65% N_2, 35% O_2/65% CO_2 (simulated RFG)	• The optimum sulfur retention temperature shifted from ~ 860 °C in enrich air combustion mode to 900~925 °C in oxy-combustion mode • About 65% of the recycled NO was reduced to N_2, and steam recirculation produced a sharp decrease in NO emission, and a slight increase in N_2O emission
Li et al. [24]	1 MWth CFB	Bituminous coal	Air, 21~50% O_2/RFG	• Under 50% O_2/RFG combustion mode, gaseous pollutant emissions in mg/MJ unit such as CO and NO are lower than that achieved by air combustion • SO_2 concentration significantly increases with levels up to three times higher compared to air combustion mode, which may produce considerably higher corrosion concerns

(continued)

Table 4.1 (continued)

Authors	Test facility	Fuels	Combustion conditions	Remarks
Moon et al [25] and Nguyen et al. [26]	100 kWth CFB	Sub-bituminous coal, lignite, wood pellet	Air, 21~27% O_2/RFG	• SO_2, NO, and CO emissions during oxy-combustion were lower than during air-combustion • With an increasing biomass co-firing ratio, SO_2, NO, and CO emissions were also reduced
Engin et al. [27]	30 kWth CFB	Lignite	Air, 21~28% O_2/RFG	• Flue gas recirculation led to a radical decrease in the emissions of NO_x (mg/MJ). Similarly, the SO_2 emission (mg/MJ) decreased with an increase in the excess oxygen ratio (EOR) in the oxy-combustion
Skopec et al. [28] and Vodička et al. [29]	30 kWth BFB	Lignite, sewage sludge, wood pellet	Air, O_2/RFG	• Oxygen staging could be used successfully to reduce NOx emissions in oxy-combustion mode • The sulfur self-retention performance of sewage sludge, and SO_2 capture efficiency of limestone both had a slight increase in oxy-fuel mode

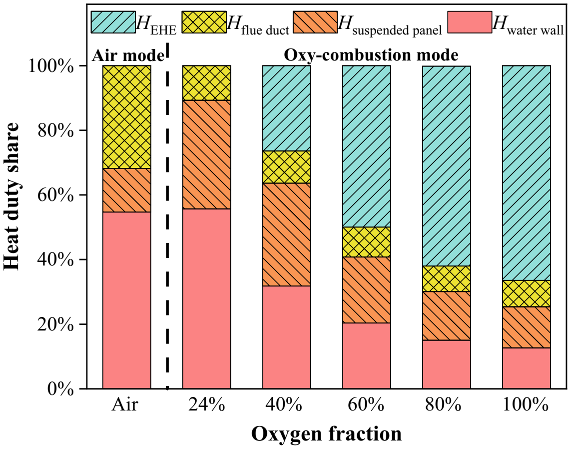
the retrofitted PC boilers. A higher O_2 concentration can reduce the impact of high CO_2 specific heat, keeping the flue gas temperature roughly consistent in both air and oxy-combustion mode, thus maintaining approximately the same heat transfer coefficient and heat distribution in both atmospheres [37]. For a retrofitted CFB boiler, it is also necessary to seriously consider the difference in fluidization in both modes. Increasing the oxygen concentration is conducive to maintaining the same average temperature of the furnace, but without changing the ratio of fuel amount and excess oxygen, it will inevitably reduce the total airflow and fluidized air velocity entering the boiler, and will bring significant direct changes to the fluidization state, particle distribution, solids recirculation fluxes, and heat balance in the fluidized bed [37, 38]. Switching between air and oxy-combustion without changes in fuel feed, heat transfer, or furnace conditions is more challenging in a CFB. Therefore, when oxy-fuel combustion is carried out in a retrofitted boiler, it is necessary to increase the fluidization velocity by increasing the fuel feeding or reducing the heat extraction in a furnace. In addition, the author of this paper also provides a feasible solution, that is, to change the particle size distribution of the bed material particles using slagging, or adding fine ash, to maintain similar particle concentration and heat transfer characteristics in the dilute phase of both modes. But for the back pass, no matter which of the above methods is adopted, due to the change of flue gas temperature, flow velocity, specific heat and radiation characteristics under oxy-fuel conditions, a separate analysis is required for each heating surface, and the heat balance between the two combustion modes is achieved by adding or short-circuiting a portion of the heating tubes [39].

For the newly designed oxy-CFB boiler, the high O_2 fraction can effectively reduce boiler size and operating costs, however, the small furnace will bring a great challenge to heat duty distribution and the layout of heating surfaces. The fluidized bed has a large amount of high-temperature bed materials, which can be further used with EHEs, making it easier to arrange the heating surfaces in CFB boilers than in PC boilers. Figure 4.3 shows that the heat load proportion of EHEs increases with O_2 fraction in the combustion atmosphere [40]. For the oxy-CFB boiler, the heating surfaces inside a furnace, including the water wall, wing and suspended panels, still occupy the dominant heat duty share in a 24% O_2/76% RFG atmosphere. When the O_2 fraction exceeds 50%, the EHEs become the most important heat extraction terms. Usually, EHEs are connected after the loop seals, and a large circulating solid flux is used to extract heat from the furnace to the EHEs [41]. However, the high circulating solids flux will also lead to an increase in the tube's wearing, so a trade-off must be made between O_2 fraction, the layout of the heating surface and the safety operation of boilers. Unlike the traditional EHEs, which only use high-temperature particles for heat exchange, a new device called an internal bed heat exchanger (IBHX) was proposed by Southeast University and Babcock & Wilcox Power Generation Group [42, 43]. The IBHX is connected to the dense phase of the furnace through some specially designed channels. The coal char burns in IBHX and heats the bed material, and then the heat is extracted from the high-temperature bed material into the high-pressure steam. The IBHX is reliant on solids internal to the furnace, so the location of the heat exchanger is not dependent on alignment with the furnace exterior walls and the outlet of a hot cyclone. According to the test results of 2.5 MWth pilot CFB, the heat absorption capacity of a single IBHX can reach 17.5–20% of total thermal input, which can effectively solve the contradiction of the heating surface layout under a high O_2 fraction.

4.5 Scale-Up and Demonstration Projects

Although a great number of lab-based studies have demonstrated the promise of oxy-combustion techniques in terms of CO_2 capture, further scale-up to industrial application still needs to overcome some technical barriers, such as the heat duty distribution in heating surfaces, and the operation strategies under different boiler loads. This necessitates studies on pilot and larger-scale fluidized bed units. Typical

Fig. 4.3 The share of different heat extraction terms normalized by the total thermal power (278 MWth, Data quoted from ref. [40])



demonstrations of oxy-fuel fluidized beds are listed in Table 4.2. It can be seen that the thermopower input of these oxy-combustion CFB facilities is mostly less than 1 MW, and only the IET, Alstom, Valmet of Finland and CIUDEN have MW-class CFB facilities. The 10 MWth (2 MWe) unit at the Korea Institute of Energy Research (KIER) is still under construction now and no further details have been reported.

CIUDEN and Foster Wheeler established the world‘s largest oxy-CFB demonstration power plant in 2012 and conducted a very comprehensive combustion test [16]. The operation time was up to 1300 h, including 920 h of the oxy-combustion test. The results show that the switching process between air-combustion mode and oxy-combustion mode is flexible. The high O_2 fraction (24~40%) tests were conducted on that platform in 2013–2014 [44], and after 14 days of continuous operation, the whole system worked smoothly with high oxygen combustion, the furnace temperature could be flexibly controlled by changing the bed material circulation rate.

In comparison to the monoethanolamine (MEA) absorption method, which is another widely used carbon capture technology in coal-fired power plants, oxy-fuel combustion has certain economic advantages, even when considering boiler retrofit, ASU & CPU equipment, and their operational costs. Vu et al. [45] conducted an economic analysis based on a 500 MWe supercritical CFB power unit and found that when employing MEA absorption and oxy-combustion for carbon capture, the electricity generation efficiency decreased from 45% to 35.5% and 38.7%, and the CO_2 avoided cost in these two scenarios was 25 and 20 €/tCO_2 respectively.

Although oxy-fuel combustion technology has certain advantages in terms of economics, it is still not as flexible in operation as the MEA absorption method. MEA absorption method only requires adding a bypass before the chimney, without causing major impacts on the existing power plant equipment and operations. In contrast, the combustion atmosphere and heat transfer characteristics have significant changes in the oxy-fuel combustion mode, thus requiring specialized research and design in these aspects.

Table 4.2 Pilot and industrial scale oxy-combustion CFB units

Institutions	Capacity of boiler	O_2 fraction at boiler inlet (%)	CO_2 concentration of flue gas (%)	Main research fields	References
Fundacion Ciudad de la Energia (CIUDEN), Spain	30 MWth	24–40	~ 80	Engineering validation; heat transfer; pollutant emissions; combustion mode switch; agglomeration and corrosion	[16, 44]
Valmet R&D Center, Finland	4 MWth	21–28	82	Engineering validation; heat transfer; pollutant emissions; combustion mode switch	[18]
Alstom Power Inc	3 MWth	30–50	> 70	Engineering validation; heat transfer; pollutant emissions; agglomeration	[46]
Southeast University, China	2.5 MWth	/	/	Heat transfer	[42, 43]
	50 kWth	21–40	93	Pollutant emissions; combustion mode switch; modeling	[15, 19]
Korea Institute of Energy Research, Korea	2 MWe	/	/	Under construction	[17, 26]
	100 kWth	21–29	91.2–95.4	Heat transfer; pollutant emissions; combustion mode switch; modeling	
Institute of Engineering Thermophysics (IET), Chinese Academy of Sciences	1 MWth	21–50	~ 90	Heat transfer; pollutant emissions; combustion mode switch	[24, 47–49]
	0.8 MWth	29	91	Pressurized oxy-combustion; combustion mode switch	

(continued)

Table 4.2 (continued)

Institutions	Capacity of boiler	O_2 fraction at boiler inlet (%)	CO_2 concentration of flue gas (%)	Main research fields	References
	100 kWth	21–55	84.1–93.9	Heat transfer; pollutant emissions; combustion mode switch; modeling	
CanmetENERGY, Canada	1 MWth	/	/	Pressurized oxy-combustion	[50, 51]
	0.8 MWth	21–29	81.8–86.9	Pollutant emissions; combustion mode switch; corrosion	[22, 52]
	100 kWth	34–45	84.4–92.9	Pollutant emissions; combustion mode switch	[21, 53, 54]
The University of Utah, USA	300 kWth	27	/	Pollutant emissions	[55]
University of Stuttgart, Germany	150 kWth	26–36	89.5	Pollutant emissions	[56]
VTT Technical Research Centre, Finland	100 kWth	24–31	70–80	Pollutant emissions; combustion mode switch; modeling	[39, 57]
Czestochowa University of Technology, Poland	100 kWth	21–35	/	Pollutant emissions; modeling	[58, 59]
University of Zaragoza, Spain	90 kWth	20–40	/	Pollutant emissions; ash deposition	[60, 61]
Tallinn University of Technology, Estonia	60 kWth	21–30	/	Pollutant emissions	[62]

4.6 Challenges and Solutions

Recent advancements in oxy-fuel combustion technology have been focused on enhancing efficiency, reducing operational costs, and addressing the challenges posed by high-temperature operations. One notable development is the improvement in oxygen production methods, such as cryogenic air separation and membrane technologies, which have become more energy-efficient and cost-effective. These advancements are crucial since oxygen production is a significant contributor to the overall energy consumption of oxy-fuel combustion systems.

Unlike the lab- and pilot-scale CFBs that usually operate in a fixed load, the industrial-scale oxy-CFB boilers must have good flexibility with a wide load range to meet the demand of peak shaving for the power grid. A feasible operation strategy is to combine an oxy-CFB boiler with an energy storage system, that is, the thermal input of the boiler remains stable, and the spare electric power is used to produce and store liquid oxygen [63, 64]. During the peak period of the power grid, turning down the output of ASU reduces its electricity consumption, and releases the stored liquid oxygen for fuel combustion. However, there is relatively rare research on another solution strategy, which is to increase the proportion of RFG and reduce the O_2 fraction of the combustion atmosphere in the low boiler load condition. The high proportion of RFG increases the amount of primary gas and gas velocity in the dense phase of the boiler, which ensures the stability of fluidization with a low load. The reduction of O_2 fraction also reduces the amount of fuel and heat input. The stability of furnace temperature and high efficiency of combustion can be well ensured under the low-load operation by reducing the circulating solids flux entering EHEs, thus facilitating the oxy-CFB boilers with a wide load regulation range.

Both theoretical analysis and demonstration results show that excessive power consumption in ASU and compression/purification unit (CPU) has become a key barrier to the commercial application of oxy-combustion technology. For a conventional coal-fired power plant, the net power efficiency will be reduced by at least 10% if it is converted to oxy-combustion mode [65]. In order to solve this problem, researchers have proposed some novel concepts like pressurized oxy-combustion and oxy-steam combustion.

Under the atmospheric oxy-combustion process, both the ASUs and CPUs are operated under high pressure, while the combustion unit is operated under atmospheric pressure, then the gas pressure fluctuation among ASU, boiler and CPU will cause the energy loss and a great penalty in the net efficiency. So, the pressurized oxy-combustion (POC) technology is proposed [66, 67]. Compared with the atmospheric system, the POC system avoids the power loss caused by pressure fluctuation. In addition, since the whole system is operated under pressure, air leakage can be effectively eliminated, the concentration of CO_2 in the flue gas can be increased significantly, and the power consumption of the CPU is also reduced. At present, this technology is still in its infancy, and due to the difficulty of high-pressure experiments, the current research is mainly in the lab-scale.

Different from the conventional oxy-combustion system, the oxy-steam combustion technology uses water vapor instead of RFG to neutralize the high temperature generated by the pure oxygen combustion of fuel [68, 69]. The energy consumption of the water circulation pump is much smaller than that of the RFG fan, which can reduce a considerable amount of energy. By analyzing the characteristics and development history of oxy-combustion CFB technology, it can be concluded that this technology has unique advantages for efficient and low-cost CO_2 capture in coal-fired power plants. The scientific research on oxy-steam combustion can effectively promote the development and industrial application of large-scale CO_2 capture technology, which is very positive and important for achieving CO_2 reduction targets.

Since its concept was proposed in the 1980s, the development of oxy-combustion fluidized bed technology has gone through the stages of laboratory experimentation, pilot testing, and industrial demonstration. This technology has evolved into one of the most promising and economically viable carbon capture technologies for large-scale coal-fired power plants. However, the high investment and operational costs still hinder its widespread promotion. Furthermore, further research is needed on the combustion mechanism, heat transfer characteristics, heat load distribution, and matching ability with the peak shaving demand of the power grid.

Oxy-fuel combustion in FBR presents a significant advancement in the field of carbon capture. However, the implementation of oxy-fuel combustion in FBRs is not without its challenges. These include high operational costs, material durability under high-T and high-O_2 conditions, and the complexities of integrating carbon capture systems. Addressing these challenges through innovative research and development is key to unlocking the full potential of this technology.

References

1. Luo W, Wang Q, Huang X, Liu Z, Zheng C (2015) Dynamic simulation and transient analysis of a 3 MWth oxy-fuel combustion system. Int J Greenhouse Gas Control 35:138–149
2. Leckner B (2022) Hundred years of fluidization for the conversion of solid fuels. Powder Technol 411:117935
3. Horn FL, Steinberg M (1982) Control of carbon dioxide emissions from a power plant (and use in enhanced oil recovery). Fuel 61:415–422
4. Andries J, Becht JGM, Hoppesteyn PDJ (1997) Pressurized fluidized bed combustion and gasification of coal using flue gas recirculation and oxygen injection. Energy Convers Manage 38:S117–S122
5. Hosoda H, Hirama T, Azuma N, Kuramoto K, Hayashi J-I, Chiba T (1998) NO_X and N_2O emission in bubbling fluidized-bed coal combustion with oxygen and recycled flue gas: macroscopic characteristics of their formation and reduction. Energy Fuels 12:102–108
6. Li K, Zeng Y, Luo J-L (2021) Corrosion performance of candidate boiler tube alloys under advanced pressurized oxy-fuel combustion conditions. Energy 215:119178
7. Li K, Zeng Y, Luo JL (2020) Condensed phase corrosion of P91 and DSS 2205 steels at advanced oxygen-fired pressurized fluidized bed combustion plants. Mater Corros 72:757–771
8. Liu Q, Zhong W, Yu A, Wang C-H (2022) Co-firing of coal and biomass under pressurized oxy-fuel combustion mode in a 10 kWth fluidized bed: nitrogen and sulfur pollutants. Chem Eng J 450:138401

9. Vasudevan S, Farooq S, Karimi IA, Saeys M, Quah MCG, Agrawal R (2016) Energy penalty estimates for CO_2 capture: comparison between fuel types and capture-combustion modes. Energy 103:709–714
10. Dobó Z, Backman M, Whitty KJ (2019) Experimental study and demonstration of pilot-scale oxy-coal combustion at elevated temperatures and pressures. Appl Energy 252:113450
11. Bu C, Pallarès D, Chen X, Gómez-Barea A, Liu D, Leckner B et al (2016) Oxy-fuel combustion of a single fuel particle in a fluidized bed: char combustion characteristics, an experimental study. Chem Eng J 287:649–656
12. Duan L, Li L, Liu D, Zhao C (2019) Fundamental study on fuel-staged oxy-fuel fluidized bed combustion. Combust Flame 206:227–238
13. Kim D, Choi S, Shaddix CR, Geier M (2014) Effect of CO_2 gasification reaction on char particle combustion in oxy-fuel conditions. Fuel 120:130–140
14. Shaddix CR, Hecht ES, Gonzalo-Tirado C, Haynes BS (2022) Quantifying the effect of CO_2 gasification on pulverized coal char oxy-fuel combustion. Proc Combust Inst 39:3379–3388
15. Duan L, Zhao C, Zhou W, Qu C, Chen X (2011) O_2/CO_2 coal combustion characteristics in a 50 kWth circulating fluidized bed. Int J Greenhouse Gas Control 5:770–776
16. Lupion M, Alvarez I, Otero P, Kuivalainen R, Lantto J, Hotta A et al (2013) 30 MWth CIUDEN oxy-CFB boiler—first experiences. Energy Procedia 37:6179–6188
17. Yang C, Kim Y, Bang B, Jeong S, Moon J, Mun T-Y et al (2020) Oxy-CFB combustion technology for use in power-generation applications. Fuel 267:117206
18. Seddighi Khavidak S, Pallarès D, Normann F, Johnsson F, Ylä-Outinen V (2015) Heat transfer in a 4 MWth circulating fluidized bed furnace operated under oxy-fired and air-fired conditions: modeling and measurements. Int J Greenhouse Gas Control 37:264–273
19. Duan L, Sun H, Zhao C, Zhou W, Chen X (2014) Coal combustion characteristics on an oxy-fuel circulating fluidized bed combustor with warm flue gas recycle. Fuel 127:47–51
20. Kim YB, Gwak YR, Keel SI, Yun JH, Lee SH (2019) Direct desulfurization of limestones under oxy-circulating fluidized bed combustion conditions. Chem Eng J 377:119650
21. Jia L, Tan Y, Wang C, Anthony EJ (2007) Experimental study of oxy-fuel combustion and sulfur capture in a mini-CFBC. Energy Fuels 21:3160–3164
22. Jia L, Tan Y, McCalden D, Wu Y, He I, Symonds R et al (2012) Commissioning of a 0.8 MWth CFBC for oxy-fuel combustion. Int J Greenhouse Gas Control 7:240–243
23. de las Obras-Loscertales M, Rufas A, de Diego LF, García-Labiano F, Gayán P, Abad A et al (2013) Effects of temperature and flue gas recycle on the SO_2 and NO_X emissions in an oxy-fuel fluidized bed combustor. Energy Procedia 37:1275–1282
24. Li H, Li S, Ren Q, Li W, Xu M, Jz L et al (2014) Experimental results for oxy-fuel combustion with high oxygen concentration in a 1 MWth pilot-scale circulating fluidized bed. Energy Procedia 63:362–371
25. Moon J-H, Jo S-H, Park SJ, Khoi NH, Seo MW, Ra HW et al (2019) Carbon dioxide purity and combustion characteristics of oxy firing compared to air firing in a pilot-scale circulating fluidized bed. Energy 166:183–192
26. Nguyen HK, Moon J-H, Jo S-H, Park SJ, Seo MW, Ra HW et al (2020) Oxy-combustion characteristics as a function of oxygen concentration and biomass co-firing ratio in a 0.1 MWth circulating fluidized bed combustion test-rig. Energy 196:117020
27. Engin B, Kayahan U, Atakül H (2020) A comparative study on the air, the oxygen-enriched air and the oxy-fuel combustion of lignites in CFB. Energy 196:117021
28. Skopec P, Hrdlička J, Vodička M (2023) Desulfurization in co-firing of sewage sludge and wooden biomass in a bubbling fluidized bed combustor under air and oxy-fuel conditions. Fuel 342:127709
29. Vodička M, Hrdlička J, Skopec P (2021) Experimental study of the NO_X reduction through the staged oxygen supply in the oxy-fuel combustion in a 30 kWth bubbling fluidized bed. Fuel 286:119343
30. Bolea I, Romeo LM, Pallarès D (2014) Heat transfer in the external heat exchanger of oxy-fuel fluidized bed boilers. Appl Therm Eng 66:75–83

31. Seddighi S (2017) Design of large scale oxy-fuel fluidized bed boilers: constant thermal power and constant furnace size scenarios. Energy 118:1286–1294
32. Cai R, Deng B, Tao X, Zhang Y, Yang H, Yue G et al (2020) Effects of horizontal tube arrays on heat transfer in an external heat exchanger. Appl Therm Eng 181:115964
33. Eder C, Hofer G, Pröll T (2021) Wall-to-bed heat transfer in bubbling fluidized bed reactors with an immersed heat exchanger and continuous particle exchange. Ind Eng Chem Res 60:7417–7428
34. Johansson R, Leckner B, Andersson K, Johnsson F (2013) Influence of particle and gas radiation in oxy-fuel combustion. Int J Heat Mass Transf 65:143–152
35. Guo J, Liu Z, Huang X, Zhang T, Luo W, Hu F et al (2017) Experimental and numerical investigations on oxy-coal combustion in a 35 MW large pilot boiler. Fuel 187:315–327
36. Fraga GC, Silva FR, Zanin LGB, da Fonseca RJC, França FHR, Centeno FR (2021) A comprehensive evaluation of the WSGG model for air- and oxy-fuel combustion conditions through three-dimensional calculations. Fire Saf J 125:103433
37. Leckner B, Gómez-Barea A (2021) Change of existing circulating fluidized bed boilers to oxy-firing conditions for CO_2 capture. Appl Energy Combust Sci 8:100042
38. Leckner B (2023) Negative CO_2 emission from oxy-fuel combustion in CFB boilers. Fuel 333:126425
39. Kuivalainen R, Pikkarainen T, Leino T, Tourunen A (2009) Development of CFB technology to provide flexible air/oxy operation for a power plant with CCS. In: The 34th international technical conference on coal utilization fuel systems
40. Seddighi S, Clough PT, Anthony EJ, Hughes RW, Lu P (2018) Scale-up challenges and opportunities for carbon capture by oxy-fuel circulating fluidized beds. Appl Energy 232:527–542
41. Blaszczuk A, Pogorzelec M, Shimizu T (2018) Heat transfer characteristics in a large-scale bubbling fluidized bed with immersed horizontal tube bundles. Energy 162:10–19
42. McCauley K, Kraft D, Maryamchik M, Wietzke D, Alexander K (2012) IR-CFB boilers: supercritical once-through developments for power generation. Power-Gen Asia
43. Maryamchik M, Wietzke D (2010) B&W IR-CFB: operating experience and new developments. In: Proceedings of the 20th international conference on fluidized bed combustion
44. Gómez M, Fernández A, Llavona I, Kuivalainen R (2014) Experiences in sulphur capture in a 30 MWth circulating fluidized bed boiler under oxy-combustion conditions. Appl Therm Eng 65:617–622
45. Vu TT, Lim Y-I, Song D, Mun T-Y, Moon J-H, Sun D et al (2020) Techno-economic analysis of ultra-supercritical power plants using air- and oxy-combustion circulating fluidized bed with and without CO_2 capture. Energy 194:116855
46. Liljedahl GN, Turek DG, Nsakala NY, Mohn NC, Fout TE (2006) Alstom's oxygenfired CFB technology development status for CO_2 mitigation. In: 31st international technical conference on coal utilization and fuel systems
47. Li S, Li H, Li W, Xu M, Eddings EG, Ren Q et al (2017) Coal combustion emission and ash formation characteristics at high oxygen concentration in a 1 MWth pilot-scale oxy-fuel circulating fluidized bed. Appl Energy 197:203–211
48. Li W, Xu M, Li S (2018) Calcium sulfation characteristics at high oxygen concentration in a 1 MWth pilot scale oxy-fuel circulating fluidized bed. Fuel Process Technol 171:192–197
49. Li W, Li S, Ren Q, Tan L, Li H, Liu J et al (2014) Study of oxy-fuel coal combustion in a 0.1 MWth circulating fluidized bed at high oxygen concentrations. Energy Fuels 28:1249–1254
50. Chowdhury M (2020) Design and operability of a pressurized oxy-combustion system. The University of Texas, El Paso
51. Yang Z, Khatri D, Verma P, Li T, Adeosun A, Kumfer BM et al (2021) Experimental study and demonstration of pilot-scale, dry feed, oxy-coal combustion under pressure. Appl Energy 285:116367
52. Tan Y, Jia L, Wu Y, Anthony EJ (2012) Experiences and results on a 0.8 MWth oxy-fuel operation pilot-scale circulating fluidized bed. Appl Energy 92:343–347
53. Wu Y, Wang C, Tan Y, Jia L, Anthony EJ (2011) Characterization of ashes from a 100 kWth pilot-scale circulating fluidized bed with oxy-fuel combustion. Appl Energy 88:2940–2948

54. Jia L, Tan Y, Anthony EJ (2009) Emissions of SO_2 and NO_X during oxy-fuel CFB combustion tests in a mini-circulating fluidized bed combustion reactor. Energy Fuels 24:910–915
55. Ahn J, Okerlund R, Fry A, Eddings EG (2011) Sulfur trioxide formation during oxy-coal combustion. Int J Greenhouse Gas Control 5:S127–S135
56. Hofbauer G, Beisheim T, Dieter H, Scheffknecht G (2014) Experiences from oxy-fuel combustion of bituminous coal in a 150 kWth circulating fluidized bed pilot facility. Energy Procedia 51:24–30
57. Seddighi KS, Pallarès D, Normann F, Johnsson F (2013) Carbon monoxide formation during oxy-fuel-fired fluidized-bed combustion. Energy Fuels 27:2275–2282
58. Krzywanski J, Czakiert T, Blaszczuk A, Rajczyk R, Muskala W, Nowak W (2015) A generalized model of SO_2 emissions from large–and small-scale CFB boilers by artificial neural network approach. Fuel Process Technol 137:66–74
59. Krzywański J, Czakiert T, Muskała W, Nowak W (2011) Modelling of CO_2, CO, SO_2, O_2 and NO_X emissions from the oxy-fuel combustion in a circulating fluidized bed. Fuel Process Technol 92:590–596
60. Lupiáñez C, Guedea I, Bolea I, Díez LI, Romeo LM (2013) Experimental study of SO_2 and NO_X emissions in fluidized bed oxy-fuel combustion. Fuel Process Technol 106:587–594
61. Lupiáñez C, Díez LI, Romeo LM (2014) Influence of gas-staging on pollutant emissions from fluidized bed oxy-firing. Chem Eng J 256:380–389
62. Konist A, Neshumayev D, Baird ZS, Anthony EJ, Maasikmets M, Jarvik O (2020) Mineral and heavy metal composition of oil shale ash from oxyfuel combustion. ACS Omega 5:32498–32506
63. Hu Y, Li X, Li H, Yan J (2013) Peak and off-peak operations of the air separation unit in oxy-coal combustion power generation systems. Appl Energy 112:747–754
64. Hanak DP, Powell D, Manovic V (2017) Techno-economic analysis of oxy-combustion coal-fired power plant with cryogenic oxygen storage. Appl Energy 191:193–203
65. Duan Y, Duan L, Anthony EJ, Zhao C (2017) Nitrogen and sulfur conversion during pressurized pyrolysis under CO_2 atmosphere in fluidized bed. Fuel 189:98–106
66. Xia F, Yang Z, Adeosun A, Gopan A, Kumfer BM, Axelbaum RL (2016) Pressurized oxy-combustion with low flue gas recycle: computational fluid dynamic simulations of radiant boilers. Fuel 181:1170–1178
67. Cao Z, Liang C, Zhou X, Zeng X, Duan L, Chen X et al (2022) Radiative heat transfer in splash and dilution zones of the pressurized oxy-fuel combustion CFB considering particle-dependent scattering. Adv Powder Technol 33:103697
68. Li Z, Zou R, Hong D, Ouyang J, Jiang L, Liu H et al (2020) Effect of CO_2 and H_2O on char properties. Part 2: in situ and ex situ char in oxy-steam combustion. Energy Fuels 34:7554–7563
69. Yue S, Wang C, Xu Z, Wang D, Zheng F, Anthony EJ (2022) The role of H_2O in NO formation and reduction during oxy-steam combustion of bituminous coal char. Combust Flame 237:111883

Open Access This chapter is licensed under the terms of the Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/), which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license and indicate if changes were made.

The images or other third party material in this chapter are included in the chapter's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the chapter's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.

Chapter 5
Chemical Looping Combustion (CLC) in FBRs

5.1 Introduction to Chemical Looping Combustion

To date, the chemical looping combustion (CLC) technology has been recognized as a highly promising CO_2 capture strategy for the combustion of fossil fuel attributed to its inherent ability of CO_2 separation (Fig. 5.1) [1]. The basic idea of CLC was first proposed in the 1950s by Lewis and Gilliland as a way of producing CO_2 with high purity, which describes a concept similar to the current known CLC process [2, 3]. Later, Richter and Knoche proposed in early 1983 a reverse reaction scheme aiming to increase the thermal efficiency in fossil fuel-fired power plants, which is pretty similar to the current CLC process [4]. Such a scheme is suggested to have two intermediate reactions using a metal oxide (copper oxide, nickel oxide, or cadmium oxide) as an oxygen carrier. The formal name of CLC was first nominated by Ishida et al. in 1987 during their thermodynamic study on natural gas-fired power plants to reduce exergy loss [5]. Seven years later, the CO_2 capture feature has been highlighted by Ishida and Jin by applying the CLC process for CO_2 capture using Fe- and Ni-based oxygen carriers [6]. However, the CLC technology was still on the documents during those days. It is worth noting that the Grangemouth Advanced CO_2 Capture Project (GRACE) was the first trial that supported the CLC technology in Europe, within which several hundred oxygen carrier materials have been investigated and a 10 kWth CLC unit has been built for the continuous operation of CLC processes [7]. Until 2011, More than 4000 h of operation experience had been achieved by successfully demonstrating the technology of CLC on combustors in the size range of 0.3–140 kWth [8]. Following these successes, CLC units from 1 to 3 MWth had been built mainly with globally collaborative efforts [9, 10].

© The Author(s) 2025

L. Duan and D. Lu, *Fluidized Bed Reactors for Carbon Capture*,
https://doi.org/10.1007/978-981-96-0274-2_5

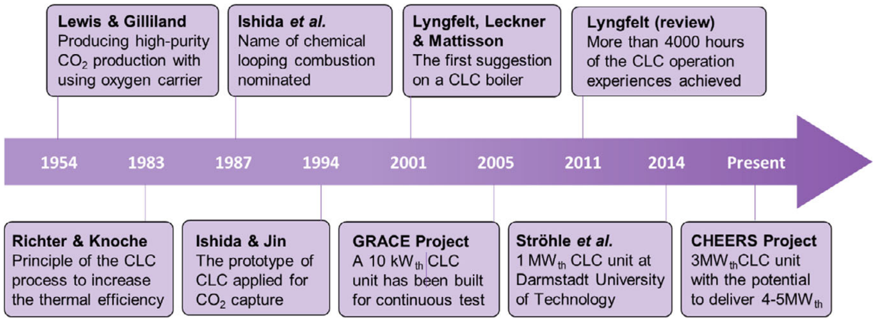


Fig. 5.1 The milestones during the development of CLC technologies [1]

5.2 Mechanics of CLC

The CLC process is ideal to be carried out with two interconnected reactors in terms of an air reactor (AR) and a fuel reactor (FR). Oxygen carriers (OCs) which are usually composed of metal oxides are circulated between two reactors in transferring oxygen from the AR to the FR (see Fig. 5.2). During this looping process, the fuel in the FR is oxidized by either the lattice oxygen of OCs or the molecular O_2 released by the OCs. Then, the O_2-deficient OC in its reduced form is transferred into the AR where it is oxidized by the air and replenished with oxygen. At the exit of the AR, the OCs are separated and returned to the FR for the next cycle. In the FR, the fuel which may be in the solid or gas phase is oxidized according to the reaction shown in Eq. 5.1:

$$(2n + m)Me_xO_y + C_nH_{2m} \leftrightarrow (2n + m)Me_xO_{y-1} + m\,H_2O + n\,CO_2 \tag{5.1}$$

In the AR, the OC is re-oxidized by the air according to the reaction shown in Eq. 5.2.

$$Me_xO_{y-1} + 1/2O_2 \leftrightarrow Me_xO_y \tag{5.2}$$

In such a reaction configuration, direct contact between the fuel and air is avoided. Thereby the exhaust stream of the FR consists mainly of CO_2 and H_2O. The H_2O can be easily condensed, resulting in a pure stream of CO_2 which can be pressurized and sequestered. The stream from the AR exit contains mainly N_2 and unreacted O_2. In the CLC technology, the conventional CO_2 capture via gas–gas separation step during or after the combustion process is avoided, which largely eliminates the energy penalty during CO_2 capture.

For large-scale continuous operation, the CLC technology is highly dependent on the availability of OC materials concerning appropriate chemical and physical properties, and more importantly, the suitable reactor system regarding the effective

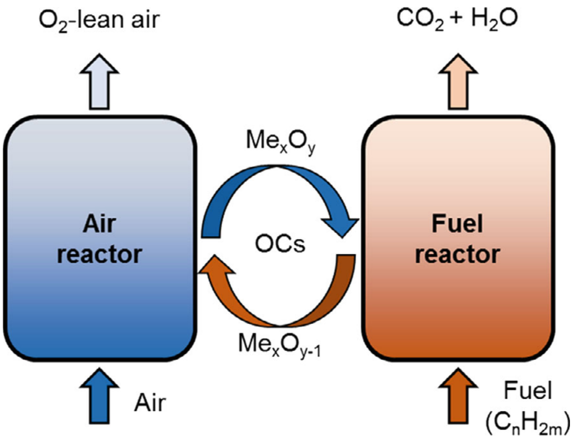


Fig. 5.2 The CLC principle: in a fuel reactor, fuel reacts with lattice oxygen of OCs, *i.e.* the metal oxide, or the molecular O_2 released by OCs in a stoichiometric ratio thereby reducing the OCs. In the air reactor, the O_2 in the air oxidizes the OCs which are subsequently recycled to the fuel reactor for a new cycle

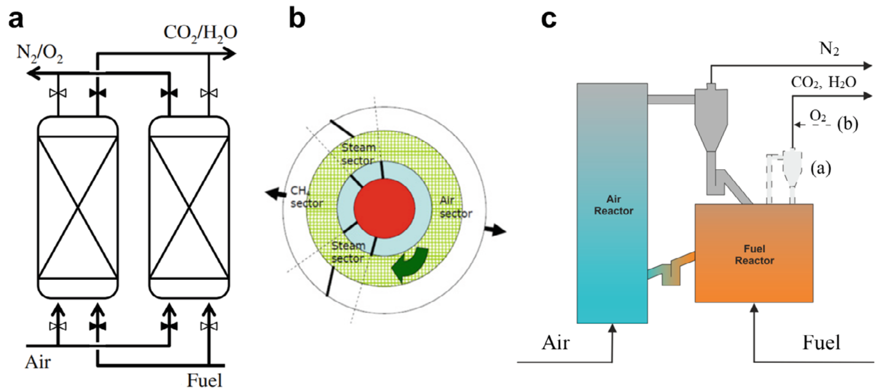


Fig. 5.3 Possible reactor concepts for chemical-looping combustion: **a** fixed or packed bed reactors; **b** rotary reactors (reprinted from ref. [18, 19] with permission of Elsevier); and **c** interconnected FBRs with (a) carbon stripper and (b) oxygen polishing equips (from ref. [20], licensed under CC-BY 4.0)

handling of a large amount of OC particles and circulating them between the FR and AR with respectively different chemical environments. Summarily, three different types of reactors have been designed to promote the CLC into practice (see Fig. 5.3), including fixed or packed bed reactors operated by a gas-switching mode [11–13], rotary reactors operated by a bed rotating [14–16], and interconnected fluidized beds operated by circulating the OCs [17].

5.3 Oxygen Carrier Materials

Theoretically, CLC for burning a gaseous fuel, such as CH_4 and syngas, is straightforward since the fuel could be easily fed to the FR and directly reacts with the lattice oxygen from OCs. Therefore, the first continuous test for CLC used gaseous fuel [21]. The major concern in the CLC for gaseous fuels is the fast decline of OC reactivity and rapid loss of OC integrity during successive redox cycles, resulting in poor fuel combustion performance and operation issues. The chemically synthesized OCs such as the Fe-, Cu- and Mn-based ones exhibit very promising reactivity, and some of them show relatively long lifetimes verified by pilot tests [22–25]. Thus, for a given fuel rate, the CLC process by using synthetic highly reactive materials requires a smaller solids inventory, which in turn leads to a smaller reactor. However, the extremely high cost of the chemically synthetic OCs associated with their sophisticated production procedure and expensive raw feedstock materials makes their usage prohibitive on large scale. The attrition, agglomeration and deactivation during the operation of a practical CLC facility always happens, which leads to a large fraction loss of the precious OCs. Another OC candidate is non-manufactured materials such as waste materials or natural ores, which are mainly represented by the red mud, iron, manganese and ilmenite ore [26]. These OCs provide pretty low material cost, while their reactivity toward gaseous fuel is much poorer than the synthetic OCs. Weak reactivity of OCs means the interconnected-fluidized beds should be operated with high circulation rates and very high solid inventory, which could be a big challenge to achieve in a practical CLC system. If the reduction reactivity of OCs could be increased while maintaining the oxygen transporting capacity, it would be possible to reduce the circulation rate of OCs. Several methods including doping and pretreatment have been used for reactivity enhancement of the ore-based OCs, while these semi-synthetic OCs are expected to be demonstrated out of the lab [27–29].

In the case of using solid fuels, such as coal and biomass, which are more abundant and cheaper than gaseous ones, the reaction between OCs and fuels becomes extremely slow or even prohibited due to the kinetic limit for solid–solid reactions [30]. To increase the reaction rate between solid fuels and OCs, two major approaches have been proposed: in situ Gasification Chemical Looping Combustion (*i*G-CLC) and Chemical Looping with Oxygen Uncoupling (CLOU) [31–33]. In the *i*G-CLC process, solid fuels with steam and/or CO_2 are introduced into the FR. During the reaction, solid fuels are firstly gasified by steam and/or CO_2 to produce CO and H_2 as intermediate gaseous feedstock. Thus, the OCs can react with the gaseous intermediates at promising reaction rates to make solid fuel combustion possible. In the *i*G-CLC, the reaction rate of gasification between solid fuels and gasifying agents is always slower than that of gas–solid reaction between OCs and volatiles or gaseous gasification products. As a result, fuel gasification is the limiting step affecting the overall fuel conversion rate in the FR. The CLOU approach can be regarded as an improvement of the conventional CLC process. The major difference between CLOU and conventional CLC processes is the mechanism for the oxidation of fuels in FR. CLOU utilizes OCs, such as the Cu- and Mn-based ones as well as metal oxides with perovskite structure, that can release gas-phase O_2 in the FR under the reducing environments with low O_2 partial pressure, and the released O_2 subsequently fully oxidizes with solid fuels. In the AR, the OCs are re-oxidized by air, which is identical to the conventional CLC process.

5.4 Design and Scale-Up Challenges

The OCs are crucial materials that link the functions of two reactors, and the proper handling of the OC materials is of utmost importance. From the operational point of view, interconnected fluidized beds are believed to have advantages over alternative designs. Firstly, the interconnected fluidized beds can easily create two or even more segregated chemical environments functioning as FR or AR [34]. Secondly, the interconnected fluidized beds which rely on the technique of fluidization excel in transferring large amounts of solids between different reactors where diverse chemical reactions can happen. Thirdly, the deployment of CFBs for power plants has the added advantage that both large (> 350 MW) atmospheric and pressurized systems also exist, and so the technical challenges of developing interconnected fluidized bed systems for a scheme of CLC operation are significantly reduced [35]. These years, considerable research attention has been paid to the CLC technology which has been demonstrated from lab and bench scale testing (< 5 kWth) to sub-pilot scale testing (5–50 kWth) until pilot scale testing (50 kWth–1 MWth), as listed in Table 5.1. Over 35 pilot reactors have been built using solid or gaseous fuels, however, most of them are based on units of interconnected fluidized beds [36–38]. It is worth mentioning that Lyngfelt A and Leckner B, have successfully designed a 1000 MWth boiler for chemical looping combustion of solid fuels, which strongly demonstrated the suitable design of the reaction systems and promising cost benefits of the CLC technology for CO_2 capture.

Moreover, from a chemical reaction point of view, using FBRs for CLC operation also exhibits obvious advantages. The fluidized bed FR shows less risk of carbon formation. The carbon formation normally happens when the OCs, *e.g.* NiO, are deeply reduced. The reduced OCs, *i.e.* metallic Ni, function as a catalyst and the carbon formation reaction initiates on its active sites. In the fluidized reactor, the rapid mixing and the fast movement of the solids make the solid components in the

Table 5.1 Representative units for CLC using interconnected fluidized beds

Centre and location	Unit (kWth)	Air reactor	Fuel reactor	Ref
Chalmers, Sweden	10	Turbulent fluidized bed and a pneumatic riser	Bubbling fluidized bed	[39]
SEU, China	10	A single riser	Spouted bed	[40]
SEU, China	1	A single riser	Spouted bed	[41]
CSIC, Spain	0.5–1.5	Bubbling fluidized bed and a pneumatic riser	Bubbling fluidized bed	[42, 43]
TU Hamburg, Germany	25	A single riser	Two-staged bubbling fluidized bed	[44]
Chalmers, Sweden	100	Turbulent fluidized bed and a pneumatic riser	Turbulent fluidized bed	[45]
TU Darmstadt, Germany	1000	A single riser	Circulating fluidized bed	[9]
SEU, China	50	A single riser	Fast fluidized bed	[46]
CSIC, Spain	50	Turbulent fluidized bed and a pneumatic riser	Turbulent fluidized bed	[47]
HUST, China	5	Turbulent fluidized bed and a pneumatic riser	Bubbling fluidized bed	[48]
HUST, China	50	Turbulent fluidized bed and a pneumatic riser	Bubbling fluidized bed	[49]
VTT, Finland	20	Turbulent fluidized bed and a pneumatic riser	Bubbling fluidized bed	[50]
SEU, China	5	A single riser	A spouted bed and a bubbling fluidized bed	[51]

whole bed highly uniform. Thus, the fuel can always encounter a portion of OCs in its oxidized state, which lowers the risk of carbon formation. While in the reactors as a fixed bed, the OCs at the bed entrance are immediately and fully converted into the reduced OCs at the beginning of the reaction, resulting in a permanent catalytic zone at the entrance and leading to severe carbon formation followed by complex side reactions. Besides, higher CO_2 selectivity can be achieved by using an FBR. An ideal reactor design should allow the whole OCs reduction period, *i.e.* the fuel oxidation period, to generate mostly CO_2 while keeping the unburnt species such as CO and CH_4 as low as possible. Due to the highly uniform solid composition within the bed, the fluidized bed can minimize the possible side reactions and thus maximize the CO_2 selectivity. Moreover, the fluidized bed can offer nearly isothermal and isobaric conditions throughout the reactor attributed to the rapid mixing of solids and the active mixing of the solids and gases. For a given reaction, great temperature fluctuations will lead to the generation of hot and cold spots where the reaction becomes strongly unstable. Thus, the uniform bed temperature in the fluidized bed guarantees stable fuel combustion in the endothermic FR and also alleviates the thermal shock on the OCs in the AR. Meanwhile, the FBR with excellent solid mixing provides uniform pressure distribution, which keeps the reaction rate nearly constant, and subsequently minimizes the risk of reaction-induced temperature fluctuations.

The challenges for operating interconnected fluidized beds mainly lay in the appropriate oxygen supply to the fuel reactor, heat generation, temperature, and pressure controls, as well as the control over global and internal solid circulation rates [20]. In particular, the following several specific criteria should be carefully considered for the design of a CLC system based on interconnected fluidized beds. Firstly, the circulation rate for OC between the FR and AR as well as the solid inventory in the respective reactors must be optimized since the rates of the reactions in FR and AR are significantly different. During CLC processes, the chemical reactions happening in different reactors are not the same, thus their reaction rates and heat-releasing behaviors are different. This imposes difficulty in the management of OC circulation between reactors. The rate of the reaction between the fuel and OC determines the amounts of OC solids needed for the FR. If the reaction rate is high enough, the amount of OC solids needed by the FR is low, otherwise, more OC solids are needed for a reaction with a low reaction rate in the FR.

Also, for a given reaction rate of the OC, more OC is needed in the FR if the oxygen-carrying capacity of the OC is low, which guarantees the transfer of sufficient oxygen from the AR to the FR. Therefore, both the solid circulation rate and the solid inventory should match the oxygen-carrying capacity of the OC and the reaction rates in the individual reactor. The reactions in the AR are always much faster than those in the FR, and the gas velocity in the AR usually provides the driving force for solid circulation. Thus, fast fluidization is normally adopted for the AR, and bubbling fluidization is chosen for the FR in order to well control the solid circulation rate, obtain a reasonable solid residence time and optimize the reactor size. The inventory of bubbling bed FR can be controlled by simply varying the bed height. Depending on the types of OC and fuels, the primary reaction happening in the FR (Eq. 5.1) can be either endothermic or exothermic. In most cases especially using CH_4 as fuel with Fe- and Ni- based OCs, the reaction in the fuel reactor is endothermic. In such a case, the OCs not only transfer the oxygen from the AR to FR but also take the heat from the former to the latter to have the FR reach a sufficient temperature. Hence, the AR is usually operated at a higher temperature than the FR.

Secondly, the release of unconverted gaseous fuels from the FR and the transfer of unconverted solid fuels from the FR to the AR during the CLC of solid fuels should be prevented in order to maintain high fuel conversion and CO_2 capture efficiency. Normally, the complete conversion of the gaseous fuels, such as volatiles and gasification products, is impossible, and the full conversion of the char in the FR is even more prohibitive due to the slow gasification reaction rate. The unconverted gaseous fuels will likely leave the FR resulting in waste. With this regard, oxygen polishing has been proposed in which pure oxygen will be introduced into the outlet of the fuel reactor to fully oxidize the unconverted gaseous fuels [52]. In some cases, the unconverted gaseous fuels will be separated from CO_2 in connection with CO_2 liquefaction and recycled to FR. Besides, the unconverted char will likely be transferred from the FR to the AR along with the OC circulation. As a result, the unconverted char will be burnt in the AR which significantly lowers the CO_2 capture efficiency. In such a situation, a carbon stripper can be introduced to separate the unconverted char particles from the OCs, and the char can be transferred back to the FR. In the carbon stripper unit, achieving an effective separation between unconverted char and OCs is based on their different fluidization behaviors. Since the density of unconverted char is always much lower than the OC, at the same time, its particle size is smaller, the char particles have much lower terminal velocities. When the gas velocity in the carbon stripper has been optimized, the elutriation rates of the unconverted char and OCs can be differentiated leading to the successful separation between them. The first carbon stripper in continuous operation has been designed to be integrated with a fuel reactor of the 10 kWth chemical-looping combustors from Chalmers University, while, it has been finally concluded to be inefficient (see Fig. 5.4a) [39, 53, 54]. Then, a newly designed carbon stripper with four chambers was installed after the FR had been implemented in the 100 kWth unit from Chalmers University, which is demonstrated to be effective for increasing fuel conversion (see Fig. 5.4b) [36].

Thirdly, how to properly handle the ash in the FBRs is a great challenge for the CLC of solid fuel. The ash of solid fuels no matter whether in *i*G-CLC or CLOU processes is always mixed with the OC before and after the ash discharging. The issues raised by the ash include contaminating the OC, bed agglomeration, corrosion, and fouling of heat-transfer surfaces and making the separation of OC harder. For the fuels containing very aggressive ash components such as K, Na, Ca, etc. fuel mix is often a good way to neutralize the danger to the boiler and minimize damage to heat-transfer surfaces and OCs. When the ash has accumulated to a critical level in the reactors, it has to be discharged. It is straightforward to discard ash and OC together from the FBRs. However, this inevitably results in a significant loss of the OC and thus a high operating cost penalty. Therefore, materials with extremely low cost are preferred for the CLC of solid fuels compared to the ones prepared by synthetic methods. Currently, the magnetically separable OCs have become a focus since they

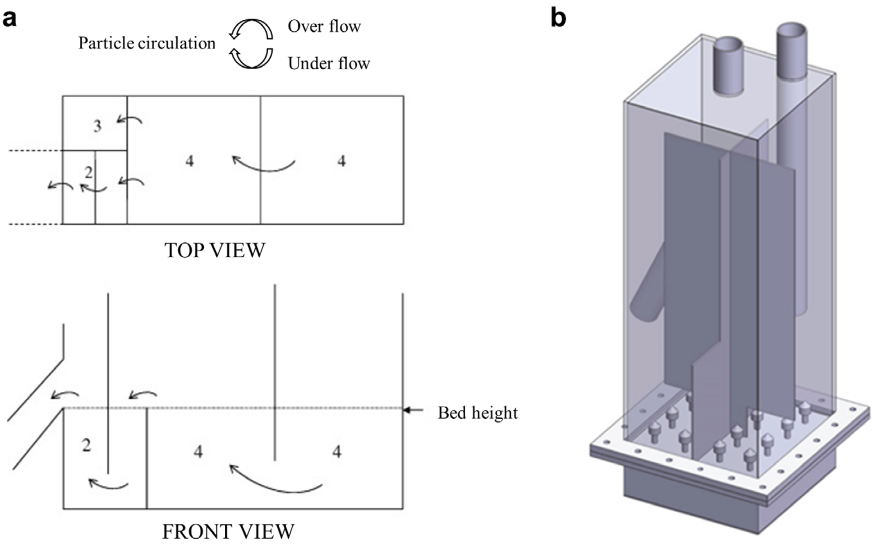


Fig. 5.4 **a** Top and front views with the particle circulation directions of the FR installed with a (2) carbon stripper of the 10 kWth chemical-looping combustors from Chalmers University (reprinted from ref. [39] with permission of Elsevier); **b** the newly designed carbon stripper with four chambers installed on the 100 kWth chemical looping unit from Chalmers University (reprinted from ref. [36] with permission of Elsevier)

hold the potential to be separated from the ash by magnetism [55]. The magnetic separation would be easier for extracting the OC from the biomass ash since it is rich in nonmagnetic oxides of Ca, Mg, Na, K, and P. However, coal ash consists of oxides of Fe exhibiting magnetic properties, which may mess up the magnetic extraction of the OCs from their ash mixture.

5.5 Case Studies and Performance Evaluation

The very pioneering reactor configuration proposed by Lewis and Gilliland along with the first invention of the CLC technique is interconnected fluidized beds with pneumatic transport lines [56]. To date, most of the experimental investigations for a continuous CLC test heavily rely on the interconnected FBR system. In such a CLC reactor system, the gas velocity in the AR provides the driving force for the solid circulation, and fast fluidization can be adopted for the AR as the reaction for OCs re-oxidation is always much faster than their reduction in the FR. The air reactor is usually designed as a riser itself or an upper riser with a bottom bed. In general, the difficulties in the design of the CLC reactor system lie in the fluidized bed of FR rather than that of AR. The major difference among the CLC systems already built based on interconnected fluidized beds is the types of FR. The frequently applied configurations chosen for the FR include a bubbling fluidized bed and a CFB.

For continuous operation, one of the most representative interconnected fluidized bed systems for CLC is the 10 kWth laboratory unit built by Chalmers University [21]. As shown in Fig. 5.5a, such a CH_4-fueled reaction system is composed of two interconnected fluidized beds which are a high-velocity riser functioning as the AR and a low-velocity bubbling fluidized bed as FR. The bed material, typically metal oxides functioning as the OCs, is circulated between the two fluidized beds. Later, this reactor system was adapted to a new unit aiming at burning solid fuels, as shown in Fig. 5.5b [39, 54]. The most important modifications to this unit are in the fuel reactor chamber with the inclusion of an additional solid recirculation loop. In the new system, the fuel reactor has been divided into three chambers: a low-velocity part where the fuel particles can be devolatilized and gasified, a carbon stripper to separate unreacted coal particles from the oxygen carrier particles, and a high-velocity part to help char particles back to the fuel reactor via a small cyclone fitted at the entrance to the fuel reactor [58].

The air reactor was a high-velocity fluidized bed connected to a riser to bring the oxygen carrier particles back to the fuel reactor. The most representative character of this Chalmers' CLC unit is using the bubbling bed as the FR, which has also been adopted by several CLC units in other research institutes such as the 500 Wth unit at CSIC and the 10 kWth unit at IFP Energies Nouvelles [42, 44]. However, several inherent issues as discussed below have been considered for the bubbling bed FRs: (1) the FR with a bubbling bed suffers from fuel gas bypassing through the bubble phase due to the coalescence of bubbles during the bubble growth, thus reducing the fuel conversion. As a result, the escaped unreacted gas has to be burned by "oxygen polishing" [59], which increases the cost and the complexity of the system; (2) the effective reactions between the gaseous gasification products and the OCs in the freeboard are rare caused by the insufficient solids holdups, leading to a poor reaction between fuel and OCs; (3) when burning solid fuels with much small particle size, the char elutriation in the bubbling bed will be serious, reducing further the fuel conversion [60].

These issues may be alleviated by increasing the solid inventory, while the increased risk of slugging and the decreased stability in fluidization will be encountered accordingly. With these regards, the 1 and 10 kWth CLC units in Southeast University have used a spout-fluidized bed as the FR instead of a bubbling bed, in which the solids mixing is intensified and the fuel residence time is prolonged. As a result, the fuel gasification reaction as well as the reaction between gasification products and OCs are intensified, while slugging in the FR has been observed due to the large solid inventory [40, 41]. To obtain a higher fuel conversion, it is worth mentioning that a two-staged bubbling bed has been adapted as the FR in a 25 kWth CLC unit at the Hamburg University of Technology (see Fig. 5.6a), which is a bit similar to the original design of the CLC system by Lewis and Gilliland [44]. In this unit, the solid fuel in the lower bed of the FR is gasified largely, and the gaseous gasification products generated from the lower bed are converted in the upper bed by the freshly oxidized OC, thus the fuel conversion has been improved. Another option

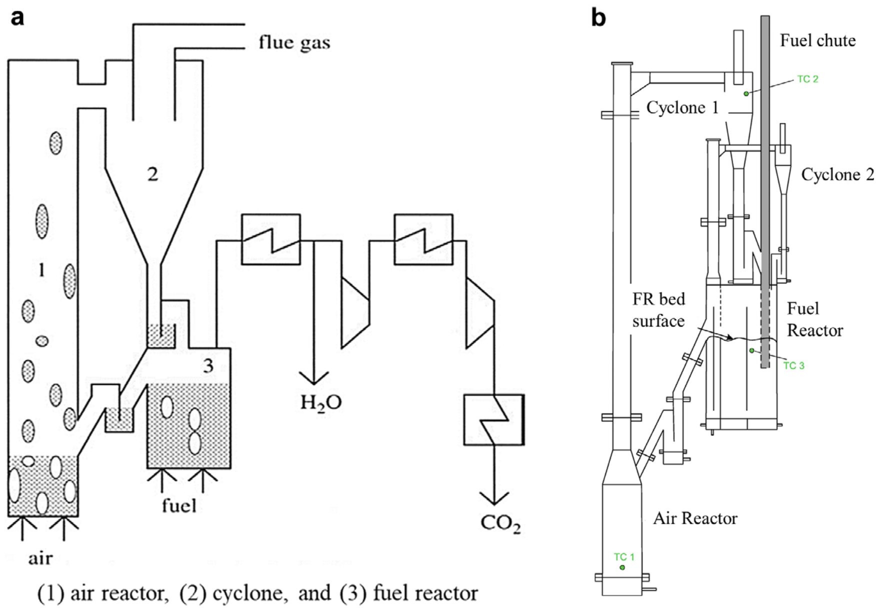


Fig. 5.5 **a** Layout of chemical-looping combustion system for burning natural gas, with two interconnected fluidized beds: (1) air reactor, (2) cyclone, and (3) fuel reactor, **b** flowsheet of the same system after a modification to burning solid fuels [21, 57, 58]

is the circulating FR with a very pioneering design proposed by Pröll et al. They have designed a riser with disturbance elements to create higher particle concentration in the riser (see Fig. 5.6b) [61].

In contrast, the CFB operated at a higher fluidization regime than the bubbling fluidized bed can provide a much better gas–solid contact along the axis of almost the whole bed, resulting in a more even and complete reaction in the reactor. Moreover, both fine and coarse particles can be effectively operated in the CFB, while the fine particles are not desirable to be used in a bubbling fluidized bed. Although the process control of a CFB is a bit more complex than the bubbling one, the scale-up potential of the former is much higher than the latter as CFB boilers are a fully proven technology for the conventional combustion of solid fuels. Therefore, a CFB has been proposed as a promising reactor configuration for the design of FR. At first, a 120 kWth CH_4-fueled bench scale CLC unit combining two circulating FBRs was built and successfully operated at the Vienna University of Technology (see Fig. 5.7a) [62]. In this system, two circulating FBRs are interconnected via a fluidized loop seal in the bottom region of the reactors (lower loop seal). The entrainment of the left-hand side primary reactor (i.e. the AR) determines global solids circulation. The solids are separated from the AR exhaust stream in a cyclone separator and pass over through a fluidized loop seal (upper loop seal) into the right-hand side secondary reactor (i.e. the FR). From there, the global solids loop closes via the lower loop seal. The FR features a circulation loop in itself (AR cyclone and internal loop seal) and is optimized with respect to good gas–solid contact and low particle attrition rates. The global circulation rate can be controlled by staged fluidization of the AR. The direct hydraulic communication of the two CFB reactors allows stable solids distribution in the system as long as the lower loop seal is designed to be large enough to not significantly hinder solids flow. Imposing moderate pressure differences between the two reactors change the theoretical solids levels in the system. This can be done by changing the backpressure from the exhaust gas lines, allowing active control of the solid hold-up in each reactor.

Later, a 1 MWth CLC pilot plant designed using a dual CFB was built at Technische Universität Darmstadt and successfully operated, which was the largest CLC unit worldwide at that time (see Fig. 5.7b) [9]. The use of a CFB as FR can decrease the necessary amounts of OCs in the system due to the better gas–solid mixing, while the residence time of the solid fuel in the FR will also be decreased compared to the FR of bubbling fluidized bed. The shorter fuel residence time is unfavorable for the char gasification reaction, i.e. the rate-limiting step of the overall fuel conversion.

5.6 Future Directions and Research Needs

After a development of more than a decade, a lot of pilot CLC units using interconnected fluidized bed systems for burning gaseous or solid fuels have been established. The successful design of an industrial-scaled CLC system is primarily related to the achievement of a stable operation between the interconnected FBRs, especially

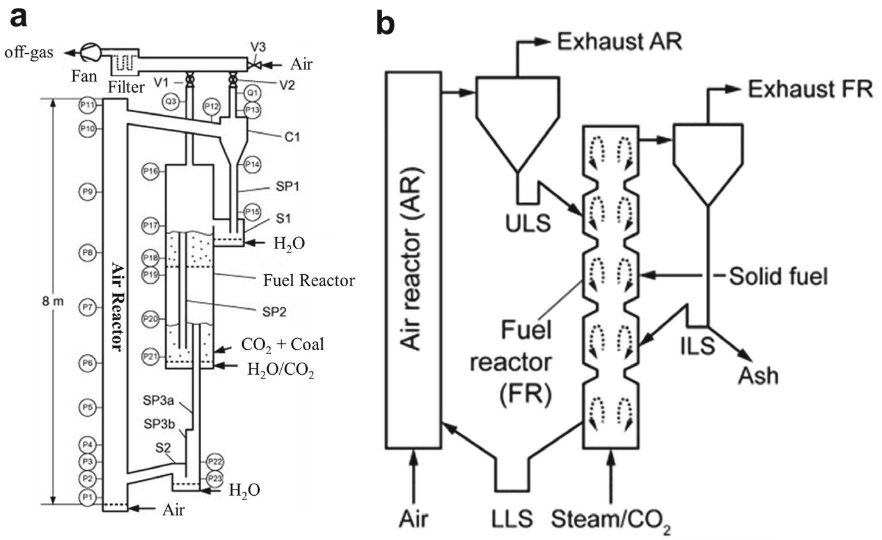


Fig. 5.6 **a** The two-staged bubbling bed of the CLC system by Thon et al. (reprinted from ref. [44] with permission of Elsevier) and **b** the newly designed fluidized reactors by Pröll et al. (reprinted from ref. [61] with permission of Elsevier)

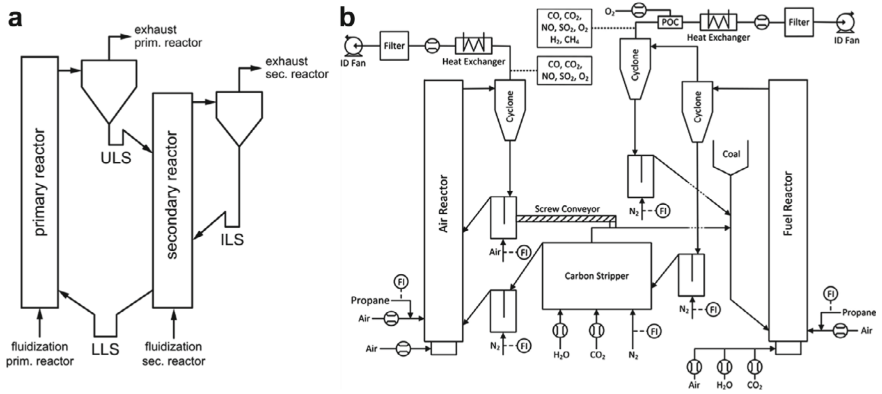


Fig. 5.7 **a** 120 kWth dual CFB CLC reactor system (LLS: 5 lower loop seal, ULS 5 upper loop seal, SR-ILS 5 secondary reactor internal loop seal) built in Vienna University of Technology (reprinted from ref. [62] with permission of Elsevier); **b** 1 MWth chemical looping plant designed with a dual CFB built-in Technische Universität Darmstadt (reprinted from ref. [9] with permission of Elsevier)

concerning load flexibility [63]. Possible challenges in the part-load operation of CLC systems include maintaining the heat balance of the system under reduced thermal input to the system and operational issues associated with the reduced gas flow rates into the reactors [64]. Therefore, a complete understanding of the dynamic behavior of interconnected fluidized bed systems is of paramount importance to successfully implement the CLC technology. Besides, the compositions of solid fuels, in practical applications, are much more complex than those of gaseous ones. A critical issue that has been seen is associated with the release of alkali metals from biomass-based solid fuels [65]. These alkali metals easily induce bed agglomeration and the OCs deactivation. Besides, the fouling and corrosion issues appear to be the biggest concerns, which require a systematical study from the mechanism to the material selection until the reactor design. Another concern in burning solid fuels is achieving a suitably high fuel conversion in the FR. This requires excellent mixing between OCs and fuels no matter the original gaseous fuels or the intermediates released from solid fuels.

In some cases, a moving bed has been adopted for FR to increase the solid residence time and to extend the chance of contact between fuel and OC, which has achieved a very smooth continuous operation in pilot scale [66]. Besides, the separation of unburned char, ashes, and OC has to be better developed in order to achieve a stable operation of the CLC system. In addition, the conversion of gas is always not complete, as a result; oxygen is required for final burnout. Thus, better contact between gas and OC, preferably in plug flow, is needed. Until now, the CLC operation on a commercial scale has not been demonstrated yet. More operational validations at a unit with a larger scale are highly desirable.

5.7 Ending Remarks

CLC is one of the most innovative technologies in CO_2 mitigation with the inherent ability of CO_2 capture when burning various regular fuels. Interconnected fluidized beds have been considered the most promising reactor for operating the CLC processes. In this chapter, the principal of CLC has been introduced and the current progress of reactor design with a special focus on the interconnected fluidized beds has been overviewed. After a development of more than a decade, although the CLC technology has been operated in various reactor systems from lab and bench scale testing to sub-pilot scale testing until pilot scale testing, it still has not been well demonstrated in the conditions close to the industrial scenario. With several case studies dealing with the CLC operation in interconnected fluidized beds, the future research directions as well as various technical concerns have been highlighted.

References

1. Chiu P-C, Ku Y (2012) Chemical looping process—a novel technology for inherent CO_2 capture. Aerosol Air Qual Res 12:1421–1432
2. Lewis WK, Gilliland ER (1954) Production of pure carbon dioxide. Patent 2665972
3. Lewis WK, Gilliland ER, Sweeney MP (1951) Gasification of carbon: metal oxides in a fluidized powder bed. Chem Eng Prog 47:251–256
4. Richter HJ, Knoche KF (1983) Reversibility of combustion processes. ACS Publications
5. Ishida M, Zheng D, Akehata T (1987) Evaluation of a chemical-looping-combustion power-generation system by graphic exergy analysis. Energy 12:147–154
6. Ishida M, Jin H (1994) A new advanced power-generation system using chemical-looping combustion. Energy 19:415–422
7. Lyngfelt A, Kronberger B, Adanez J, Morin JX, Hurst P (2005) The grace project: development of oxygen carrier particles for chemical-looping combustion. Design and operation of a 10 kW chemical-looping combustor. Elsevier Science Ltd., Oxford
8. Lyngfelt A (2011) Oxygen carriers for chemical looping combustion-4 000 h of operational experience. Oil Gas Sci Technol-Revued'IFP Energies nouvelles 66:161–172
9. Ströhle J, Orth M, Epple B (2014) Design and operation of a 1 MWth chemical looping plant. Appl Energy 113:1490–1495
10. Di Giuliano A, Capone S, Anatone M, Gallucci K (2022) Chemical looping combustion and gasification: a review and a focus on European research projects. Ind Eng Chem Res 61:14403–14432
11. Noorman S, van Sint AM, Kuipers (2007) Packed bed reactor technology for chemical-looping combustion. Ind Eng Chem Res 46:4212–4220
12. Fernández J, Alarcón J (2015) Chemical looping combustion process in fixed-bed reactors using ilmenite as oxygen carrier: conceptual design and operation strategy. Chem Eng J 264:797–806
13. Spallina V, Marinello B, Gallucci F, Romano MC, Annaland MVS (2017) Chemical looping reforming in packed-bed reactors: modelling, experimental validation and large-scale reactor design. Fuel Process Technol 156:156–170
14. Håkonsen SF, Blom R (2011) Chemical looping combustion in a rotating bed reactor–finding optimal process conditions for prototype reactor. Environ Sci Technol 45:9619–9626
15. Zhao Z, Iloeje CO, Chen T, Ghoniem AF (2014) Design of a rotary reactor for chemical-looping combustion. Part 1: fundamentals and design methodology. Fuel 121:327–343
16. Iloeje CO, Zhao Z, Ghoniem AF (2018) Design and techno-economic optimization of a rotary chemical looping combustion power plant with CO_2 capture. Appl Energy 231:1179–1190
17. Kolbitsch P, Bolhàr-Nordenkampf J, Pröll T, Hofbauer H (2010) Operating experience with chemical looping combustion in a 120 kW dual circulating fluidized bed (DCFB) unit. Int J Greenhouse Gas Control 4:180–185
18. Dahl IM, Bakken E, Larring Y, Spjelkavik AI, Håkonsen SF, Blom R (2009) On the development of novel reactor concepts for chemical looping combustion. Energy Procedia 1:1513–1519
19. Adanez J, Abad A, Garcia-Labiano F, Gayan P, De Diego LF (2012) Progress in chemical-looping combustion and reforming technologies. Prog Energy Combust Sci 38:215–282
20. Czakiert T, Krzywanski J, Zylka A, Nowak W (2022) Chemical looping combustion: a brief overview. Energies 15:1563
21. Lyngfelt A, Thunman H (2005) Construction and 100 h of operational experience of a 10-kW chemical-looping combustor. Carbon Dioxide Capture Storage Deep Geol Form-Results CO_2 Capture Proj 1:625–645
22. Cabello A, Gayán P, Pans MÁ, Dueso C, García-Labiano F, Abad A et al (2013) Evaluation of a highly reactive and sulfur resistant synthetic Fe-based oxygen carrier for CLC using gaseous fuels. Energy Procedia 37:580–587
23. Cabello A, Abad A, Gayan P, de Diego LF, Garcia-Labiano F, Adanez J (2014) Effect of operating conditions and H_2S presence on the performance of $CaMg_{0.1}Mn_{0.9}O_3-\delta$ perovskite material in chemical looping combustion (CLC). Energy Fuels 28:1262–1274

24. Penthor S, Zerobin F, Mayer K, Pröll T, Hofbauer H (2015) Investigation of the performance of a copper based oxygen carrier for chemical looping combustion in a 120 kW pilot plant for gaseous fuels. Appl Energy 145:52–59
25. Siriwardane R, Riley J, Bayham S, Straub D, Tian H, Weber J et al (2018) 50-kWth methane/air chemical looping combustion tests with commercially prepared CuO-Fe_2O_3-alumina oxygen carrier with two different techniques. Appl Energy 213:92–99
26. Leion H, Lyngfelt A, Johansson M, Jerndal E, Mattisson T (2008) The use of ilmenite as an oxygen carrier in chemical-looping combustion. Chem Eng Res Des 86:1017–1026
27. Sun Z, Lu DY, Hughes RW, Filippou D (2018) O_2 uncoupling behaviour of ilmenite and manganese-modified ilmenite as oxygen carriers. Fuel Process Technol 169:15–23
28. Sun Z, Lu DY, Ridha FN, Hughes RW, Filippou D (2017) Enhanced performance of ilmenite modified by CeO_2, ZrO_2, NiO, and Mn_2O_3 as oxygen carriers in chemical looping combustion. Appl Energy 195:303–315
29. Jinhua B, Zhenshan L, Ningsheng C (2013) Promoting the reduction reactivity of ilmenite by introducing foreign ions in chemical looping combustion. Ind Eng Chem Res 52:6119–6128
30. Chen L, Bao J, Kong L, Combs M, Nikolic HS, Fan Z et al (2016) The direct solid-solid reaction between coal char and iron-based oxygen carrier and its contribution to solid-fueled chemical looping combustion. Appl Energy 184:9–18
31. Mattisson T, Lyngfelt A, Leion H (2009) Chemical-looping with oxygen uncoupling for combustion of solid fuels. Int J Greenhouse Gas Control 3:11–19
32. Fan L-S, Zeng L, Wang W, Luo S (2012) Chemical looping processes for CO_2 capture and carbonaceous fuel conversion–prospect and opportunity. Energy Environ Sci 5:7254–7280
33. Mendiara T, Adánez-Rubio I, Gayán P, Abad A, De Diego L, García-Labiano F et al (2016) Process comparison for biomass combustion: in situ gasification-chemical looping combustion (iG-CLC) versus chemical looping with oxygen uncoupling (CLOU). Energ Technol 4:1130–1136
34. Salvador C, Lu D, Anthony EJ, Abanades J (2003) Enhancement of CaO for CO_2 capture in an FBC environment. Chem Eng J 96:187–195
35. Manovic V, Anthony EJ (2010) Lime-based sorbents for high-temperature CO_2 capture—a review of sorbent modification methods. Int J Environ Res Public Health 7:3129–3140
36. Markström P, Linderholm C, Lyngfelt A (2013) Chemical-looping combustion of solid fuels—Design and operation of a 100 kW unit with bituminous coal. Int J Greenhouse Gas Control 15:150–162
37. Mattisson T, Keller M, Linderholm C, Moldenhauer P, Rydén M, Leion H et al (2018) Chemical-looping technologies using circulating fluidized bed systems: status of development. Fuel Process Technol 172:1–12
38. Wei G, He F, Huang Z, Zheng A, Zhao K, Li H (2015) Continuous operation of a 10 kWth chemical looping integrated fluidized bed reactor for gasifying biomass using an iron-based oxygen carrier. Energy Fuels 29:233–241
39. Berguerand N, Lyngfelt A (2008) Design and operation of a 10 kWth chemical-looping combustor for solid fuels—testing with South African coal. Fuel 87:2713–2726
40. Shen L, Wu J, Gao Z, Xiao J (2009) Reactivity deterioration of NiO/Al_2O_3 oxygen carrier for chemical looping combustion of coal in a 10 kWth reactor. Combust Flame 156:1377–1385
41. Shen L, Wu J, Gao Z, Xiao J (2010) Characterization of chemical looping combustion of coal in a 1 kWth reactor with a nickel-based oxygen carrier. Combust Flame 157:934–942
42. Cuadrat A, Abad A, García-Labiano F, Gayán P, de Diego LF, Adánez J (2011) The use of ilmenite as oxygen-carrier in a 500 Wth chemical-looping coal combustion unit. Int J Greenhouse Gas Control 5:1630–1642
43. Abad A, Adánez-Rubio I, Gayán P, García-Labiano F, de Diego LF, Adánez J (2012) Demonstration of chemical-looping with oxygen uncoupling (CLOU) process in a 1.5 kWth continuously operating unit using a Cu-based oxygen-carrier. Int J Greenhouse Gas Control 6:189–200
44. Thon A, Kramp M, Hartge E-U, Heinrich S, Werther J (2014) Operational experience with a system of coupled fluidized beds for chemical looping combustion of solid fuels using ilmenite as oxygen carrier. Appl Energy 118:309–317

45. Markström P, Lyngfelt A (2012) Designing and operating a cold-flow model of a 100 kW chemical-looping combustor. Powder Technol 222:182–192
46. Xiao R, Chen L, Saha C, Zhang S, Bhattacharya S (2012) Pressurized chemical-looping combustion of coal using an iron ore as oxygen carrier in a pilot-scale unit. Int J Greenhouse Gas Control 10:363–373
47. Adánez J, Abad A, Perez-Vega R, de Diego LF, García-Labiano F, Gayán P (2014) Design and operation of a coal-fired 50 kWth chemical looping combustor. Energy Procedia 63:63–72
48. Ma J, Zhao H, Tian X, Wei Y, Rajendran S, Zhang Y et al (2015) Chemical looping combustion of coal in a 5 kWth interconnected fluidized bed reactor using hematite as oxygen carrier. Appl Energy 157:304–313
49. Ma J, Tian X, Wang C, Chen X, Zhao H (2018) Performance of a 50 kWth coal-fuelled chemical looping combustor. Int J Greenhouse Gas Control 75:98–106
50. Pikkarainen T, Hiltunen I, Teir S (2016) Piloting of bio-CLC for BECCS. In: 4th international conference on chemical looping
51. Yan J, Shen L, Jiang S, Wu J, Shen T, Song T (2017) Combustion performance of sewage sludge in a novel CLC system with a two-stage fuel reactor. Energy Fuels 31:12570–12581
52. Lyngfelt A, Leckner B (2015) A 1000 MWth boiler for chemical-looping combustion of solid fuels—discussion of design and costs. Appl Energy 157:475–487
53. Sun H, Cheng M, Chen D, Xu L, Li Z, Cai N (2015) Experimental study of a carbon stripper in solid fuel chemical looping combustion. Ind Eng Chem Res 54:8743–8753
54. Berguerand N, Lyngfelt A (2008) The use of petroleum coke as fuel in a 10 kWth chemical-looping combustor. Int J Greenhouse Gas Control 2:169–179
55. Adánez-Rubio I, Bautista H, Izquierdo MT, Gayán P, Abad A, Adánez J (2021) Development of a magnetic Cu-based oxygen carrier for the chemical looping with oxygen uncoupling (CLOU) process. Fuel Process Technol 218:106836
56. Arnold C (1950) Process for production of carbon monoxide and hydrogen. Pat Specification 636:206
57. Lyngfelt A, Leckner B, Mattisson T (2001) A fluidized-bed combustion process with inherent CO_2 separation: application of chemical-looping combustion. Chem Eng Sci 56:3101–3113
58. Schmitz M, Linderholm C (2016) Performance of calcium manganate as oxygen carrier in chemical looping combustion of biochar in a 10 kW pilot. Appl Energy 169:729–737
59. Mei D, Linderholm C, Lyngfelt A (2021) Performance of an oxy-polishing step in the 100 kWth chemical looping combustion prototype. Chem Eng J 409:128202
60. Song T, Shen L (2018) Review of reactor for chemical looping combustion of solid fuels. Int J Greenhouse Gas Control 76:92–110
61. Penthor S, Stollhof M, Pröll T, Hofbauer H (2016) Detailed fluid dynamic investigations of a novel fuel reactor concept for chemical looping combustion of solid fuels. Powder Technol 287:61–69
62. Stollhof M, Penthor S, Mayer K, Hofbauer H (2019) Influence of the loop seal fluidization on the operation of a fluidized bed reactor system. Powder Technol 352:422–435
63. Spinelli M, Peltola P, Bischi A, Ritvanen J, Hyppänen T, Romano MC (2016) Process integration of chemical looping combustion with oxygen uncoupling in a coal-fired power plant. Energy 103:646–659
64. Peltola P, Alobaid F, Tynjälä T, Ritvanen J (2022) Overview of fluidized bed reactor modeling for chemical looping combustion: status and research needs. Energy Fuels 36:9385–9409
65. Gogolev I, Linderholm C, Gall D, Schmitz M, Mattisson T, Pettersson JBC et al (2019) Chemical-looping combustion in a 100 kW unit using a mixture of synthetic and natural oxygen carriers—operational results and fate of biomass fuel alkali. Int J Greenhouse Gas Control 88:371–382
66. Zhang Y, Wang D, Pottimurthy Y, Kong F, Hsieh T-L, Sakadjian B et al (2021) Coal direct chemical looping process: 250 kW pilot-scale testing for power generation and carbon capture. Appl Energy 282:116065

Open Access This chapter is licensed under the terms of the Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/), which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license and indicate if changes were made.

The images or other third party material in this chapter are included in the chapter's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the chapter's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.

Chapter 6
Calcium Looping in FBRs

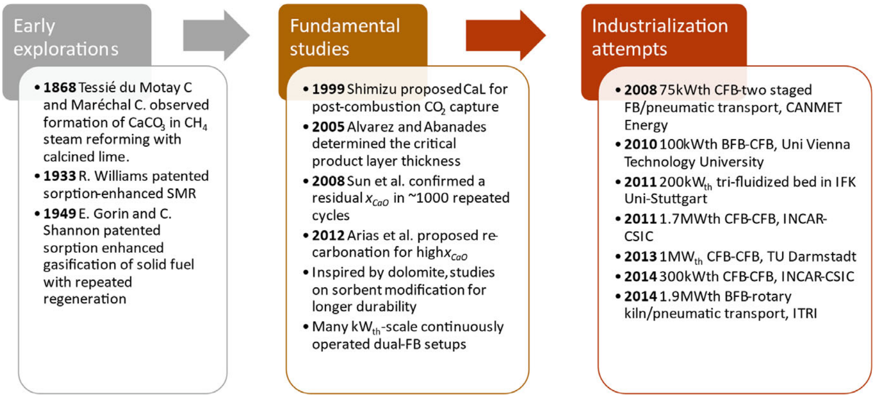
6.1 Introduction to Calcium Looping

The calcium looping process, an emerging technology in carbon capture, hinges on the reversible reaction of calcium oxide (CaO) with carbon dioxide to form calcium carbonate ($CaCO_3$) [1]. This process occurs in two primary stages: the carbonation stage, where CaO reacts with CO_2 to capture it as $CaCO_3$ (Eq. 6.1), and the calcination stage, where the $CaCO_3$ is heated to release the captured CO_2 and regenerate the CaO sorbent (reversed Eq. 6.1). The ability of calcium looping to effectively capture CO_2 from large-scale emissions sources, such as power plants and industrial facilities, makes it a promising technology for combating climate change [2]. Figure 6.1 illustrates the roadmap for the development of the calcium looping technology.

$$CaO + CO_2 = CaCO_3 \tag{6.1}$$

The use of CaO to absorb CO_2 can be dated back to the eighteenth century while the concept of Calcium Looping (CaL), i.e., repeatedly absorbing CO_2 by CaO, was investigated around the 1950s when Gorin and Shannon [3] patented a process that separates CO_2 in the coal-gasification gas in order to generate purer hydrogen. This process was not commercialized because of its low competitiveness. CaL attracted more attention due to the need to remove CO_2 from the flue gas. Shimizu and coworkers described this process in a relatively complete form [4]. CaL has attracted more attention since and then became a well-documented efficient technique for CO_2 capture from flue gas. Later, many pilot- to demonstration-scale (i.e., from 1 kWth up to 1.9 MWth) units were built, with attempts to industrialize this process. The development of CaL is summarized in Fig. 6.1 and Table 6.1. CANMET Energy reported the first demonstration of CaL with continuous sorbent looping, by using a 75 kWth dual fluidized bed system composed of an oxy-fired CFB calciner and a bubbling fluidized bed carbonator [5]. The University of Stuttgart successfully demonstrated the CaL process in a 10 kWth lab-scale dual fluidized bed unit, then

© The Author(s) 2025
L. Duan and D. Lu, *Fluidized Bed Reactors for Carbon Capture*,
https://doi.org/10.1007/978-981-96-0274-2_6



- **1868** Tessié du Motay C and Maréchal C. observed formation of $CaCO_3$ in CH_4 steam reforming with calcined lime.
- **1933** R. Williams patented sorption-enhanced SMR
- **1949** E. Gorin and C. Shannon patented sorption enhanced gasification of solid fuel with repeated regeneration

- **1999** Shimizu proposed CaL for post-combustion CO_2 capture
- **2005** Alvarez and Abanades determined the critical product layer thickness
- **2008** Sun et al. confirmed a residual X_{CaO} in ~1000 repeated cycles
- **2012** Arias et al. proposed re-carbonation for high X_{CaO}
- Inspired by dolomite, studies on sorbent modification for longer durability
- Many kW_{th}-scale continuously operated dual-FB setups

- **2008** 75kWth CFB-two staged FB/pneumatic transport, CANMET Energy
- **2010** 100kWth BFB-CFB, Uni Vienna Technology University
- **2011** 200kW_{th} tri-fluidized bed in IFK Uni-Stuttgart
- **2011** 1.7MWth CFB-CFB, INCAR-CSIC
- **2013** 1MW_{th} CFB-CFB, TU Darmstadt
- **2014** 300kWth CFB-CFB, INCAR-CSIC
- **2014** 1.9MWth BFB-rotary kiln/pneumatic transport, ITRI

Fig. 6.1 Development of calcium looping

upscaled the system to a 200 kWth three fluidized bed facilities [6, 7]. The CaL process, comprising a turbulent fluidized bed carbonator and a CFB regenerator, demonstrated excellent flexibility under both high and low plant capacity factors. The design of such a 200 kWth pilot-scale unit maximizes the operating flexibility and allows operating at the realistic CO_2 capture condition. TU-Darmstadt built a 1 MWth pilot plant using two interconnected CFB reactors. A CO_2 capture efficiency of higher than 80% was achieved under various operating conditions for more than 2000 h operation [8]. On the grounds of the design and operation experience gained from a 30 kWth bench-scale unit for CaL process [5], the Instituto Nacional del Carbón—Consejo Superior de Investigaciones Científica (INCAR-CSIC) successfully developed a pilot-scale 1.7 MWth plant that comprises two interconnected CFB reactors [9]. A CO_2 capture of more than 90% has been demonstrated in the 1.7 MWth plant operated under conditions close to a real-scale unit [10]. Industrial Technology Research Institute (ITRI) designed a 1.9 MWth CaL pilot plant that includes a rotary kiln calcinatory and demonstrated the feasibility of integrating the CaL process into cement production [11]. After 300 h of continuous operation in a cement manufacturing plant, such a CaL system achieved a CO_2 capture efficiency of 85% as well as a capture rate of higher than 1 tCO_2/h.

6.2 Process Fundamentals

As shown in Fig. 6.2, the CaL process typically employs dual-FBRs for the regenerator and carbonator to continuously conduct the calcination and carbonation reaction to cofacilitate an in-situ CO_2 capture from flue gas [2]. In general, CaL is a temperature swing process. During each CaL cycle, the calcination of calcium carbonate is firstly conducted at temperatures above 900 °C with its energy supplied by oxy-fuel combustion, leading to the generation of a CO_2-rich flue gas stream ready for

Table 6.1 Representative bench- and pilot-scale FBRs for the CaL process

Research institute	Year	Size (kWth)	Carbonator				Regenerator			
			Type[a]	Diameter (m)	Height (m)	Temperature (°C)	Type[a]	Diameter (m)	Height (m)	Temperature (°C)
Industrial Technology Research Institute	2011 [41]	1	BFB	0.1	2.5	600–700	MB	0.05	0.9	800–900
	2013 [41]	3	BFB	0.1	2.5	600–700	RK	0.27	5	500–1000
	2013 [11]	1900	BFB	3.3	4.2	650	RK	0.9	5	500–1000
INCAR-CSIC	2011 [42]	30	CFB	0.1	6.5	568–722	CFB	0.1	6	800–1000
	2011 [43]	1700	CFB	0.65	15	600–715	CFB	0.75	15	820–950
Darmstadt University of Technology	2014 [26]	1000	CFB	0.59	8.66	650–670	CFB	0.4	11.35	< 1000
University of Stuttgart	2011 [6]	10	BFB	0.114	3.5	630–700	CFB	0.071	12.4	850–900
	2011 [44]	200	CFB	0.023	10	650	CFB	0.021	10	875–930
	2014 [7]	200	CFB	0.033	6	600–680	CFB	0.021	10	875–930
Vienna University of Technology	2010 [45]	100	BFB	0.28	2	650	CFB	0.08	5	850
CANMET Energy	2008 [5]	75	BFB/MB	0.1	2–5	580–720	CFB	0.1	4.5–5	850–950
Cranfield University	2014 [9]	25	EB	0.1	4.3	600–650	BFB	0.615	1.2	900–950
Tsinghua University	2009 [10]	10	BFB	0.149	1	630	BFB	0.117	1	850

[a] BFB: bubbling fluidized bed; CFB: CFB; MB: moving bed; EB: entrained bed; RK: rotary kiln

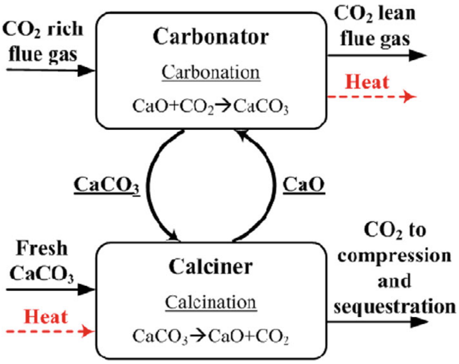


Fig. 6.2 Schematic diagram of the calcium looping process (from ref. [13], licensed under CC-BY 4.0)

sequences. The calcined CaO-based sorbent is then used for CO_2 capture at a temperature around 650 °C in the carbonator, which produces CO_2-lean flue gas after CO_2 capture. Carbonation has been noted with a rapid initial rate, followed by a remarkable decline to a very slow reaction rate due to the inefficient mass transfer and heat transfer resistance associated with the yield of $CaCO_3$ product layer [12]. In contrast, calcination was demonstrated to complete rapidly in minutes. Particle size of limestone, partial pressure of CO_2, and operation temperature were documented as important factors that largely affect the kinetics of both reactions.

6.3 Sorbent Development

Sorbent deactivation over cycles manifests an inevitable issue of CaL. As a result of sintering, sulphation, and attrition, sorbent deactivation leads to a remarkable decrease in CO_2 capture efficiency. Previous studies showed that the conversion rate of CaO in the limestone abruptly decreased to ~ 0.15 within ~ 20 cycles [14]. To counteract the performance decay of sorbents, two typical strategies can be summarized: (1) pretreatment of natural limestones, and (2) the development of synthetic sorbents. A make-up stream is widely used to refresh the spent sorbents, which has been demonstrated to enhance sorbent cyclic reactivity by increasing porosity. However, the friable nature of hydrated sorbents can negate the merit of reactivation because they could break up into fine particles, which could be easily entrained out of the FBRs. Thermal pretreatment manifests another widely-used strategy to improve the reactivity of natural limestone. Previous research [15] has proved that thermally pretreated limestone particles could retain high reactivity over up to a thousand cycles. Nonetheless, the attrition resistance of the particles was markedly reduced. Though MgO intrinsically shows lower CO_2 uptake than CaO, doping pretreatment of Ca-based sorbents by dolomites and magnesites has been suggested to exhibit superior long-term CO_2 reactivity [2]. The incorporation of MgO can not only maintain the porosity during the carbonation process, but also lead to less sintering of particles because of the higher melting point of MgO. Synthetic sorbents are widely investigated with higher CO_2 uptake and faster reaction kinetics than that of natural limestone owing to their high surface areas and favorable pore size distributions. In general, synthetic sorbents were prepared by dispersing CaO across an inert matrix, such as mayenite and γ-Al_2O_3. Pacciani et al. [16] found the synthetic CaO/mayenite showed stable porosity throughout the cycling test. Florin and Harris [17] attributed the faster CO_2 capture kinetics of synthetic sorbents to a solid-state diffusion mechanism. Despite that, synthetic sorbents cannot get rid of the attrition problem when applying them in CFBRs. Pelletization of synthetic materials has been demonstrated to alleviate this problem. By using calcium aluminate cement as binders, existing work manifested the promise of palletization to improve the attrition resistance of synthetic sorbents [18].

6.4 Economic Viability and Environmental Benefits

In comparison with oxy-fuel combustion and amine scrubbing systems, CaL technology reveals remarkable advantages in terms of efficiency penalty and environmental benefits. Regarding the oxy-fuel power plant, the net efficiency would be largely reduced by 8–12% points, mainly applied to the O_2 production through cryogenic air separation20 [19]. In contrast, for the CaL process, although oxy-fuel combustion has to be used in the regenerator, only some portion of the fuel is burned in an O_2-rich environment. As estimated by Shimazu et al., the calciner burns about 45% of the total fuel [4]. As a result, 30–50% less O_2 is required for oxy-fuel combustion in the calciner of CaL system in contrast to the oxy-fuel power plant, thus leading to a smaller size of the air separation unit and less O_2 leakage risk. The projected efficiency penalty of the CaL process accounts for 2.6–7.9% points, which is also significantly lower than the amine scrubbing system if reference MEA solvent is employed (9.5–12.5% points) [20]. In addition, in contrast to low-temperature CO_2 capture processes, the heat for temperature swing in CaL can be easily recovered to produce high-pressure steam for electricity generation [21]. Concerning CaO sorbents for CO_2 capture, natural sources like limestones and dolomite are cheap in price and globally available. More than that, $CaCO_3$ and CaO are much more friendly to the equipment and environment than amine-based solvents, which may cause equipment corrosion and environmental disposal issues. The average operating cost of CO_2 avoided by the CaL process is estimated to be \$29–50/tCO_2, exhibiting higher economic viability than that by amine scrubbing (\$50–74/tCO_2) [22] and Oxy-fuel combustion (\$35–72/tCO_2) [23].

6.5 Operational Insights and Case Studies

Operational parameters play a pivotal role in the efficiency of calcium looping in FBRs. Key parameters include temperature, pressure, gas flow rates, and the residence time of the sorbent in both the carbonation and calcination stages. Optimizing these parameters is essential to maximize CO_2 capture while minimizing energy consumption and sorbent attrition. The selection of operational parameters was found in an awkward dilemma, wherein the CO_2 capture performance was restricted by the thermodynamic limitations, while the CaL process efficiency should be maximized and the reactor size minimized simultaneously. The first issue is to regulate the reactor temperature within the proper range under realistic operating conditions, which act as a determining factor on the operation stability and CO_2 capture efficiency of the CaL process. In general, the low-end temperature of the calciner is determined by the desired partial pressure of the CO_2 stream, while the high-end operation temperature of the calciner is limited by the sorbent sintering performance. Increasing temperature in calciner could attain high combustion and calcination efficiency, while excess higher temperature would also lead to larger heat demand and sorbent sintering problems. Flue gas recirculation was in general used to regulate the temperature.

The control and optimization of these operational parameters are facilitated by advanced process control systems. These systems can adjust the operating conditions in real time based on the feedstock characteristics, sorbent performance, and desired CO_2 capture efficiency. Moreover, the integration of real-time monitoring and data analytics tools can help in predicting the behavior of the system under varying conditions, enabling proactive adjustments and minimizing operational disruptions.

Another operational aspect is the selection and preparation of calcium-based sorbents. The performance of calcium looping significantly depends on the reactivity and stability of the sorbents. Ideal sorbents should exhibit high CO_2 capture capacity, rapid reaction kinetics, and durability over multiple carbonation and calcination cycles. Research into various forms of calcium oxide, including naturally occurring limestone and synthetic sorbents, is crucial to identify the most effective materials for FBR applications.

ITRI implemented this technique in the 1.9 MWth CaL pilot plant, which resulted in a more uniform temperature distribution in the calciner, thus increasing the usable length for calcination [24]. INCAR-CSIC introduced removable cooling bayonet tubes into the 1.7 MWth CaL pilot plant to regulate the temperature carbonator [25]. Based on the experimental results conducted in the 200 MWth CaL pilot plant, as shown in Fig. 6.3, the University of Stuttgart proved that a staged oxidant supply was able to provide smooth temperature profiles in the CFB calciner [7].

Another important concern is the manipulation of solid inventories between CaL reactors. An experimental campaign by ITRI in the 3 kWth CaL pilot plant highlighted that low solid inventory was detrimental to CO_2 capture efficiency while high solid inventories would require more heat supply to the regenerator, thus increasing the operation cost [11]. As reported by the 30 kWth bench-scale CaL unit developed by INCAR-CSIC, solid inventories and looping rates were regulated by the

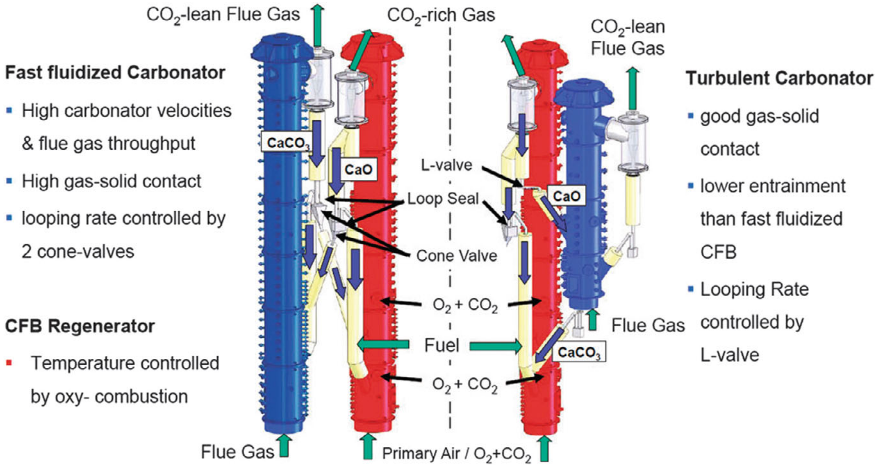


Fig. 6.3 200 kWth DFB pilot plant configurations at the University of Stuttgart with staged oxidant supply (reprinted from ref. [7] with permission of Elsevier)

high-efficiency cyclones in synergy with BFB loop seals. However, a loss of solid inventories was found due to the insufficient cyclone separation efficiency, therefore, risers of both the regenerator and carbonator were further extended to attain the dynamic looping operation. The Darmstadt University of Technology incorporated screw conveyors in the 1 MWth pilot plant to manipulate the solid transfer between two interconnected CFB reactors (see Fig. 6.4), which was said to control the solid circulation rate accurately and flexibly under different load conditions [26].

By doing so, flue gas and solid circulation between reactors were decoupled, thus facilitating the operation of the CaL system to cater to the fluctuation of flue gas loads. Similarly, the University of Stuttgart deployed the screw conveyor in synergy with loop seals in the 200 kWth pilot plant to control the solid looping rate. Such screw conveyors, however, could be mechanically impractical considering large solid loads in commercial scale reactors [20]. Varying inlet gas velocity of the carbonator was well-documented and exhibited an appreciable impact on the particle entrainment rate and the stability of dense solid region. To maintain the hydrodynamics of carbonator and CO$_2$ capture efficiency at high inlet gas velocity, recirculating solid to the bed bottom and staging flue gas at different bed heights should be further implemented. Moreover, the supply of large heat flow to the calciner is still a challenging task to achieve the desired calcination efficiency. For the case of the oxy-fuel combustor, sufficient solid inventories and circulation rates are necessary to prevent the formation of hot spots. Namely, proper control of solid inventories of industrial-scale CaL system would be confronted with more challenges and energy penalties.

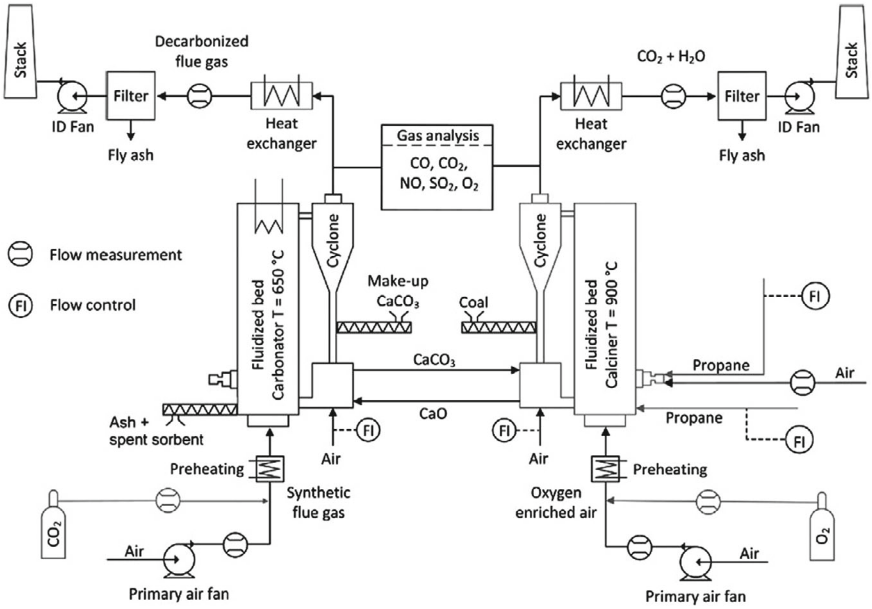


Fig. 6.4 Schematic diagram of the 1 MWth CaL system at Darmstadt University of Technology (reprinted from ref. [26] with permission of Elsevier)

6.6 Challenges and Prospective Solutions

Because of sintering, attrition and sulphation under temperature swing and cyclic operation, sorbent deactivation remains the primary challenge of CaL technology. As highlighted in Sect. 5.3, extensive studies have been conducted to improve the CO_2 capture capacity and cyclic stability of the CaO-based sorbent, mainly through (1) the reactivation and modification of natural calcium-based materials and (2) the development of synthetic sorbents [27]. However, a majority of those works lack consideration of evaluating the feasibility of the calcium-based sorbents under industry-relevant operating conditions. For instance, most of the sorbent modifications are complex and expensive, which offsets the important advantage of CaL, low cost. On the other hand, based on the performance assessment by fixed-bed thermogravimetric analyzer (TGA), CaO hollow microspheres with highly porous shells seem to possess desirable cyclic CO_2 uptake and reaction kinetics. Those CaO particles are, however, inherently difficult to fluidize due to the fine diameters (i.e., < 5 μm) [28]. Hence, their potential and feasibility for CO_2 capture in FBR require further investigation under realist operating conditions. Another critical issue is the attrition resistance of sorbent, which essentially affects the long-term CO_2 capture performance when the CaL process is scaled up to a full-size reactor. Hence, there remains much scope in the design of cheap and reactive sorbent with high attrition resistance.

By far, most of the scale-up efforts towards the CaL process were made to achieve the long-term continuous operation with desired CO_2 capture efficiency at a larger size of circulating fluidized reactors, as listed in Table 4. In contrast, the operational parameters should be carefully optimized, as to reduce the cost of the CaL process to an acceptable level. In addition, as the flue gas volume at 900 K is 3 times that at 300 K, the reactor volume of CaL is typically larger than the low temperature processes to allow appropriate residence time of the gas in the reactor. Hence, an appropriate carbonator that is reasonably sized and allows sufficient CO_2 capture efficiency is required. The latter requires sufficient gas residence time, meaning a large reactor. Oxy-fuel CFB combustion has been the primary choice to supply the heat required by the calcination, which accounts for 35~50% of the whole energy input of a CaL power plant [29]. Therefore, technical measures to improve the cost-efficiency are mainly directed at (1) reducing heat requirements in the calciner, (2) alleviating the energy penalty of the oxy-fuel CFB combustion; and (3) integrating calcium looping with other process cost-effective system. It is well-accepted that increasing the CO_2 carrying capacity of highly cycled sorbents can reduce the make-up flow demand in the CaL system, thereby lowering energy requirements and operating costs. Recarbonation is a promising approach to enhancing CO_2 carrying capacities of cycled sorbents, which has been successfully demonstrated in La Pereda 1.7 MWth CaL pilot plant [30]. As shown in Fig. 6.5, the carbonator loop seal was retrofitted to work as a recarbonator. With the aid of the recarbonator, the carrying capacity of the sorbents was remarkably improved by up to 10%. For the sake of reducing the heat demand in the calciner, Arias et al. [31] proposed a calciner configuration that affords the operation under extreme oxy-fuel conditions, thus avoiding or reducing the recycling of flue gas. As demonstrated in a 2.0 MWth CaL pilot plant, extremely high oxygen concentrations of 75% in the calciner were achieved without the observation of hot spots. Based on a similar strategy, the possibility of using pure O_2 in the calciner was experimentally demonstrated in the 25 kWth CaL plant at Cranfield University [32]. The use of pure O_2 could noticeably improve the cost efficiency because of the less energy requirement for preheating the gas steam, the less oxygen supply demand for ASU, and the smaller size of the calciner [32]. In contrast to the direct heat supply by oxy-combustion of fuel in the calciner, alternative CaL configurations are emerging to reduce the energy penalty, which transfers indirect heat sources into calciner, for instance, by using combustion flue gas as the heat source [33], using the circulated solids as the heat carriers [34], or using high-temperature heat pipes [35]. However, these indirect heating options are still at the conceptual stage due to the extra arrangement of heating surface, the high cost of heat transfer media, and the increased complexity of the whole CaL system.

Prior to scaling up FBRs into commercial CaL applications, process modeling was typically necessary to estimate the hydrodynamic feasibility, the expected CO_2 capture performance, the net energy efficiency, as well as the operation cost. However, the establishment of a reliable model for the CaL plant is sophisticated and currently underdeveloped due to the lack of reasonable assumptions, such as solid looping rates, sorbent attrition rates, ash accumulation behaviors, carbonation and calcination kinetics, etc. Most predictive models follow the hydrodynamic formulations

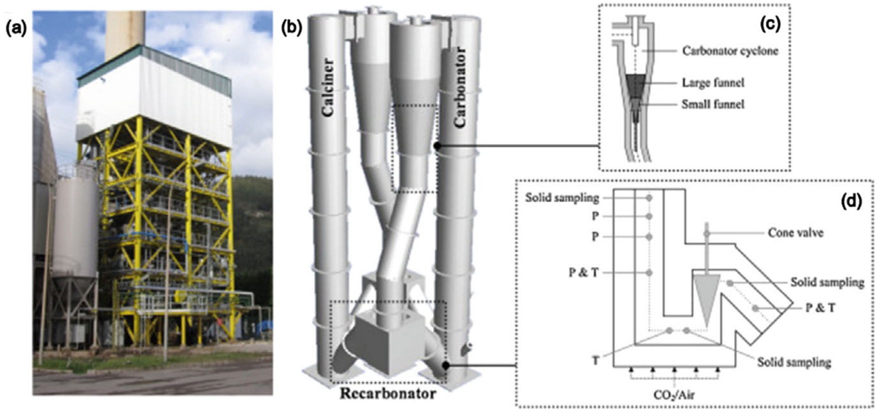


Fig. 6.5 **a** La Pereda 1.7 MWth CaL pilot plant; **b** Location of the calciner, carbonator, and recarbonator (retrofitted by the loop seal); **c** Schematic of the funnels; **d** Schematic of the recarbonator (reprinted from ref. [30] with permission of Elsevier)

proposed by Kunii and Levenspiel (K-L) [36]. For a fluidized bed with higher superficial gas velocity, the K-L CFB model, which assumes the bed is composed of a bottom dense bed and a gradually leaned bed with a near-wall dense zone and core lean zone, has been applied for many processes, and satisfactory predictions were achieved [37, 38] However, because of the lack of experimental validations, empirical parameters in the K-L model should be specified, with caution to make particular assumptions in consideration of reaction kinetics and hydrodynamics in the carbonator/calciner. Experimental parameters concerning reaction kinetics can be obtained easily by a fixed-bed TGA. Despite that, conventional TGA intrinsically suffers from severe mass transfer limitations [39]. A recent work by Li et al. [40] incorporated a micro-fluidized bed reactor (MFBR) with TGA to evaluate the carbonation kinetics. They further found that the carbonation kinetics of CaO obtained from MFB-TGA was significantly faster compared with that via conventional TGA, which implies that the carbonation and calcination kinetics evaluated by TGA should be handled cautiously when being further adapted for CaL process modeling. Beyond that, CaL process modeling requires further attempts at sorbent deactivation by including the effect of attrition, sintering, and sulphation, which are hardly reported by existing studies. In other words, the CaL community should also report more valid experimental tests or make operation information more accessible to the public, thus supporting the development of a reliable process model for the scale-up of the CaL system.

6.7 Ending Remarks

CaL technology employs FBRs, that has been well proven a promising method for in-situ CO_2 capture from flue gas. In this chapter, the process fundamentals of CaL were first introduced in terms of thermodynamics and kinetics of the reversible reactions between carbonation and calcination. The current progress of implementing CaL in FBRs was reviewed and highlighted from technical and operational insights. Despite that the CaL process has been demonstrated from lab scale to pilot scale, spanning from kWth to MWth level, plenty of challenges have to be overcome for the industrial scale-up, for instance, to mitigate sorbent deactivation, reduce operation cost, optimize process model and so on. Further endeavors should be paid in these directions to promote the CaL technology into commercialization.

References

1. Chen J, Duan L, Sun Z (2020) Review on the development of sorbents for calcium looping. Energy Fuels 34:7806–7836
2. Blamey J, Anthony E, Wang J, Fennell P (2010) The calcium looping cycle for large-scale CO_2 capture. Prog Energy Combust Sci 36:260–279
3. Everett G (1953) Gasification of carbonaceous solid fuels: United States, 2654661A
4. Shimizu T, Hirama T, Hosoda H, Kitano K, Inagaki M, Tejima K (1999) A twin fluid-bed reactor for removal of CO_2 from combustion processes. Chem Eng Res Des 77:62–68
5. Lu DY, Hughes RW, Anthony EJ (2008) Ca-based sorbent looping combustion for CO_2 capture in pilot-scale dual fluidized beds. Fuel Process Technol 89:1386–1395
6. Rodríguez N, Alonso M, Abanades JC, Charitos A, Hawthorne C, Scheffknecht G et al (2011) Comparison of experimental results from three dual fluidized bed test facilities capturing CO_2 with CaO. Energy Procedia 4:393–401
7. Dieter H, Bidwe AR, Varela-Duelli G, Charitos A, Hawthorne C, Scheffknecht G (2014) Development of the calcium looping CO_2 capture technology from lab to pilot scale at IFK, University of Stuttgart. Fuel 127:23–37
8. Arias B, Diego ME, Mendez A, Abanades JC, Diaz L, Lorenzo M et al (2017) Operating experience in La Pereda 1.7 MWth calcium looping pilot. In: 13th international conference on greenhouse gas control technologies, Ghgt-13
9. Cotton A, Finney K, Patchigolla K, Eatwell-Hall R, Oakey J, Swithenbank J et al (2014) Quantification of trace element emissions from low-carbon emission energy sources: (I) Ca-looping cycle for post-combustion CO_2 capture and (II) fixed bed, air blown down-draft gasifier. Chem Eng Sci 107:13–29
10. Fang F, Li Z, Cai N (2009) Continuous CO_2 capture from flue gases using a dual fluidized bed reactor with calcium-based sorbent. Ind Eng Chem Res 48:11140–11147
11. Chang MH, Huang CM, Liu WH, Chen WC, Cheng JY, Chen W et al (2013) Design and experimental investigation of calcium looping process for 3 kWth and 1.9 MWth facilities. Chem Eng Technol 36:1525–1532
12. Sun P, Grace JR, Lim CJ, Anthony EJ (2007) The effect of CaO sintering on cyclic CO_2 capture in energy systems. AIChE J 53:2432–2442
13. Chou Y, Chen W, Hsu H, Wan H (2018) Sorbent regeneration by cascade cyclone calciner in a pilot-scale calcium looping carbon capture system. Int J Environ Sci Dev 9:131–135
14. Erans M, Manovic V, Anthony EJ (2016) Calcium looping sorbents for CO_2 capture. Appl Energy 180:722–742

15. Chen Z, Song HS, Portillo M, Lim CJ, Grace JR, Anthony E (2009) Long-term calcination/carbonation cycling and thermal pretreatment for CO_2 capture by limestone and dolomite. Energy Fuels 23:1437–1444
16. Pacciani R, Müller CR, Davidson J, Dennis J, Hayhurst AN (2008) Synthetic Ca-based solid sorbents suitable for capturing CO_2 in a fluidized bed. Can J Chem Eng 86:356–366
17. Florin NH, Harris AT (2008) Screening CaO-based sorbents for CO_2 capture in biomass gasifiers. Energy Fuels 22:2734–2742
18. Manovic V, Anthony EJ (2009) Screening of binders for pelletization of CaO-based sorbents for CO_2 capture. Energy Fuels 23:4797–4804
19. Boot-Handford ME, Abanades JC, Anthony EJ, Blunt MJ, Brandani S, Mac Dowell N et al (2014) Carbon capture and storage update. Energy Environ Sci 7:130–189
20. Hanak DP, Anthony EJ, Manovic V (2015) A review of developments in pilot-plant testing and modelling of calcium looping process for CO_2 capture from power generation systems. Energy Environ Sci 8:2199–2249
21. Martínez I, Murillo R, Grasa G, Abanades JC (2011) Integration of a Ca looping system for CO_2 capture in existing power plants. AIChE J 57:2599–2607
22. Panja P, McPherson B, Deo M (2022) Techno-economic analysis of amine-based CO_2 capture technology: hunter plant case study. Carbon Capture Sci Technol 3:100041
23. Finkenrath M (2012) Cost and performance of carbon dioxide capture from power generation. Chem Eng Technol 35:482–488
24. Chang M, Chen W, Huang C, Liu W, Chou Y, Chang W et al (2014) Design and experimental testing of a 1.9 MW calcium looping pilot plant. In: 12th international conference on greenhouse gas control technologies, Ghgt-12
25. Arias B, Diego M, Abanades J, Lorenzo M, Diaz L, Martínez D et al (2013) Demonstration of steady state CO_2 capture in a 1.7 MWth calcium looping pilot. Int J Greenhouse Gas Control 18:237–245
26. Ströhle J, Junk M, Kremer J, Galloy A, Epple B (2014) Carbonate looping experiments in a 1 MWth pilot plant and model validation. Fuel 127:13–22
27. Gao W, Liang S, Wang R, Jiang Q, Zhang Y, Zheng Q et al (2020) Industrial carbon dioxide capture and utilization: state of the art and future challenges. Chem Soc Rev 49:8584–8686
28. Chen J, Duan L, Sun Z (2019) Accurate control of cage-like CaO hollow microspheres for enhanced CO_2 capture in calcium looping via a template-assisted synthesis approach. Environ Sci Technol 53:2249–2259
29. Rodriguez N, Alonso M, Grasa G, Abanades JC (2008) Heat requirements in a calciner of $CaCO_3$ integrated in a CO_2 capture system using CaO. Chem Eng J 138:148–154
30. Diego ME, Arias B, Méndez A, Lorenzo M, Díaz L, Sánchez-Biezma A et al (2016) Experimental testing of a sorbent reactivation process in La Pereda 1.7 MW calcium looping pilot plant. Int J Greenhouse Gas Control 50:14–22
31. Arias B, Diego M, Méndez A, Alonso M, Abanades J (2018) Calcium looping performance under extreme oxy-fuel combustion conditions in the calciner. Fuel 222:711–717
32. Erans M, Jeremias M, Zheng L, Yao JG, Blamey J, Manovic V et al (2018) Pilot testing of enhanced sorbents for calcium looping with cement production. Appl Energy 225:392–401
33. Martínez I, Murillo R, Grasa G, Rodríguez N, Abanades JC (2011) Conceptual design of a three fluidised beds combustion system capturing CO_2 with CaO. Int J Greenhouse Gas Control 5:498–504
34. Abanades JC, Anthony EJ, Wang JS, Oakey JE (2005) Fluidized bed combustion systems integrating CO_2 capture with CaO. Environ Sci Technol 39:2861–2866
35. Reitz M, Junk M, Ströhle J, Epple B (2016) Design and operation of a 300 kW indirectly heated carbonate looping pilot plant. Int J Greenhouse Gas Control 54:272–281
36. Kunii D, Levenspiel O (2000) The KL reactor model for circulating fluidized beds. Chem Eng Sci 55:4563–4570
37. Leckner B (2017) Regimes of large-scale fluidized beds for solid fuel conversion. Powder Technol 308:362–367

38. Romano MC (2012) Modeling the carbonator of a Ca-looping process for CO_2 capture from power plant flue gas. Chem Eng Sci 69:257–269
39. Schulze S, Nikrityuk P, Abosteif Z, Guhl S, Richter A, Meyer B (2017) Heat and mass transfer within thermogravimetric analyser: from simulation to improved estimation of kinetic data for char gasification. Fuel 187:338–348
40. Li Y, Li Z, Wang H, Cai N (2020) CaO carbonation kinetics determined using micro-fluidized bed thermogravimetric analysis. Fuel 264:116823
41. Huang L, Cao D (2013) Selective adsorption of olefin-paraffin on diamond-like frameworks: diamondyne and PAF-302. J Mater Chem A 1:9433–9439
42. Rodríguez N, Alonso M, Abanades J (2011) Experimental investigation of a circulating fluidized-bed reactor to capture CO_2 with CaO. AIChE J 57:1356–1366
43. Sánchez-Biezma A, Ballesteros JC, Diaz L, De Zárraga E, Álvarez FJ, López J et al (2011) Postcombustion CO_2 capture with CaO. Status of the technology and next steps towards large scale demonstration. Energy Procedia 4:852–859
44. Hawthorne C, Dieter H, Bidwe A, Schuster A, Scheffknecht G, Unterberger S et al (2011) CO_2 capture with CaO in a 200 kWth dual fluidized bed pilot plant. Energy Procedia 4:441–448
45. Aigner I, Pfeifer C, Hofbauer H (2011) Co-gasification of coal and wood in a dual fluidized bed gasifier. Fuel 90:2404–2412

Open Access This chapter is licensed under the terms of the Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/), which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license and indicate if changes were made.

The images or other third party material in this chapter are included in the chapter's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the chapter's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.

Chapter 7
Low-Temperature Sorption in FBRs

7.1 Introduction to Low-Temperature Sorption

For the CO_2 capture from low-temperature exhaust gas, two technical routes were generally used: (1) physisorption-based CO_2 capture by using porous materials; and (2) chemisorption-based CO_2 capture using solid alkaline sorbents.

Physisorption inherently relies on intermolecular interactions, especially van der Waals and electrostatic forces, between the CO_2 molecule and the sorbents. Due to the weak physical interaction between the CO_2 molecule and the sorbents, physisorption-based CO_2 capture is less sensitive to temperature and technically suitable for high partial pressure CO_2, which is not particularly applicable for CO_2 capture from post-combustion flue gas. In this case, by using fixed-bed reactors the pressure swing adsorption (PSA) strategy is preferred [1].

In chemisorption, a chemical reaction takes place between CO_2 and the sorbents that comprise various amine groups or alkaline carbonates impregnated on porous supports, such as SiO_2 and Al_2O_3. Consequently, the low-temperature CO_2 capture chemisorption-based technique involves two reactors, that is, one reactor for CO_2 capture while the other for regenerating spent sorbents. Unlike the CaL process, both reactors require a smaller unit size due to the lower operating temperature, i.e., 50–80 °C for CO_2 capture and 100–200 °C for regeneration [2]. Besides, in contrast to physisorption-based CO_2 capture, the high enthalpies involved in the chemisorption process make CO_2 chemisorption more sensitive to temperature variation, operated in a relatively narrow temperature range.

The temperature swing adsorption (TSA) process is widely adopted in CO_2 chemisorption [3]. As a result, chemisorption-based CO_2 capture can handle efficient CO_2 capture at low partial pressures, for instance, from coal-fired flue gas or even from ambient air conditions. However, heat integration and management remain a crucial concern in this process. In virtue of the enhanced heat transfer and the uniform temperature profile, fluidized bed configurations are particularly appealing to the heating and cooling stage involved in the TSA process [4]. Hence, low-temperature

© The Author(s) 2025

L. Duan and D. Lu, *Fluidized Bed Reactors for Carbon Capture*,
https://doi.org/10.1007/978-981-96-0274-2_7

CO_2 sorption processes integrated with FBRs are specifically highlighted in this section.

7.2 Sorbents for Low-Temperature Applications

Over recent decades, there has been increasing interest in developing sorbents for low-temperature CO_2 sorption, including solid amine-based materials, alkali carbonates, zeolites, porous carbons, metal–organic frameworks, and porous organic polymers [3]. Applying low-temperature CO_2 sorption in FBR requires sorbents with desired performance, for example, cheap price, high capacities at low CO_2 partial pressure (10–15%), facile regeneration temperature, high chemical and mechanical stability, etc. To date, only solid amine-based and alkali carbonates-based sorbents have been investigated to demonstrate low-temperature CO_2 sorption concepts in pilot-scale FBRs, as indicated in Table 7.1.

To improve the CO_2 adsorption capacity as well as the CO_2 selectivity, solid amine-based sorbents (SABS) were typically fabricated by supporting amines on porous materials. (SABS) like polyethyleneimine (PEI) interact with CO_2 by amino groups intensively in a chemisorption manner, thereby demanding a higher heat penalty to regenerate [5]. Another concern of (SABS) is their poor chemical stability, which is vulnerable to deterioration by SO_2 and O_2 in the flue gas. Moreover, (SABS) also exhibits a high potential risk of equipment corrosion. Given these drawbacks, their industrial widespread application is still far from fulfillment. In contrast, alkali metal carbonates attract increasing focus due to their low cost, high CO_2 uptake, desirable thermal/chemical stability, and less detrimental to the operation safety.

Alkali metal carbonates react with CO_2 to form hydrogen carbonates ($M_2CO_3 + H_2O + CO_2 \leftrightarrow 2MHCO_3, \quad M = Na/K$). K^+ ions intrinsically exhibit a stronger water affinity and a higher diffusion rate compared to Na^+ ions [6]. As a result, K_2CO_3 performs higher CO_2 uptake and faster CO_2 sorption kinetics in comparison with Na_2CO_3. To alleviate the attrition issue as well as to improve the CO_2 sorption capacity, porous supports such as Al_2O_3, SiO_2, and TiO_2 are widely used to disperse alkali metal carbonates and to increase mechanical strength. In a study using a low-temperature CO_2 sorption test in a dual FBR, Zhao et al. [7] demonstrated that the K_2CO_3/Al_2O_3 sorbent showed high CO_2 capture rate ($> 90\%$), stable porosity, and excellent attrition resistance during the 10 sorption–desorption cycles.

7.3 Process Integration and Optimization

Similar to other CO_2 capture technologies discussed above, the most important concern of low-temperature CO_2 sorptions is minimizing the energy penalty and operation costs. In pursuit of this goal, various efforts have been made by developing high-performing sorbent with high CO_2 capacity, facile regeneration at mild

Table 7.1 Demonstrations of low-temperature CO_2 sorption in FBRs

Research institute	Year	Sorbent	Scale	Reactor	CO_2 capture rate (%)	Adsorption temperature (°C)	Regeneration temperature (°C)	Regeneration penalty (MJth/kg)	Capital cost	Ref
KIER	2014	K_2CO_3	~ 200 tCO_2/d (10 MWe)	Fluidized bed	80	40–80	140–200	4.0–5.0	–	[17]
ADA	2010	Solid amine	1 MWe	Multi-staged fluidized bed	90	40	120	4.75	124–256 \$/ tCO_2	[11]
RTI International	2016	PEI/Silica	~ 0.15 tCO_2/d	Multi-staged fluidized bed	90	60–70	110–120	2.45–2.75	38–46 €/ tCO_2	[10]
TU Wien	2015	Solid amine	~ 1 tCO_2/d	Multi-staged fluidized bed	90	75	120	–	–	[12]

conditions, and cyclic stability. Beyond the sorbent performance, efficient process integration is also pivotal to minimize the energy penalty, for example, by improving CO_2 capture efficiency and managing heat requirements.

Maximizing the CO_2 capacity in the adsorption reactor is a primary concern. In a conventional FBR with a single-stage configuration, the absorbent particles are mostly well-mixed and uniformly loaded with CO_2, leading to a quick CO_2 breakthrough because of the significant increase of equilibrium CO_2 partial pressure [9]. In response to this challenge, the development of multi-stage fluidized beds has become a benchmark in current research (see Fig. 7.1a) [8]. Compared with the single-stage FBR, the multi-stage can gain benefits similar to fixed beds by reducing the internal back mixing. Wherein the sorbent particles flow counter-current to the feeding gas, which enables fresh sorbents to flow downward the bed reactor with the decreasing CO_2 partial pressure (see Fig. 7.1b). [1] This configuration provides a remarkable driving force for CO_2 adsorption, thereby improving CO_2 capture efficiency greatly at each stage.

Given these advantages, many pilot-scale studies have currently been conducted to evaluate the multi-stage fluidized bed for adsorption-based CO_2 capture [10–12]. The improved CO_2 capture efficiency by the multi-staged configuration further leads to a significant reduction in the sorbent recirculation rate, thus reducing the operation cost. To control the gas–solid interactions and maximize the CO_2 capture rate in the FBR, Ma and co-workers [13] from Southeast University proposed a bubbling-transport FBR (see Fig. 7.2a) by integrating both the bubbling and fast fluidized

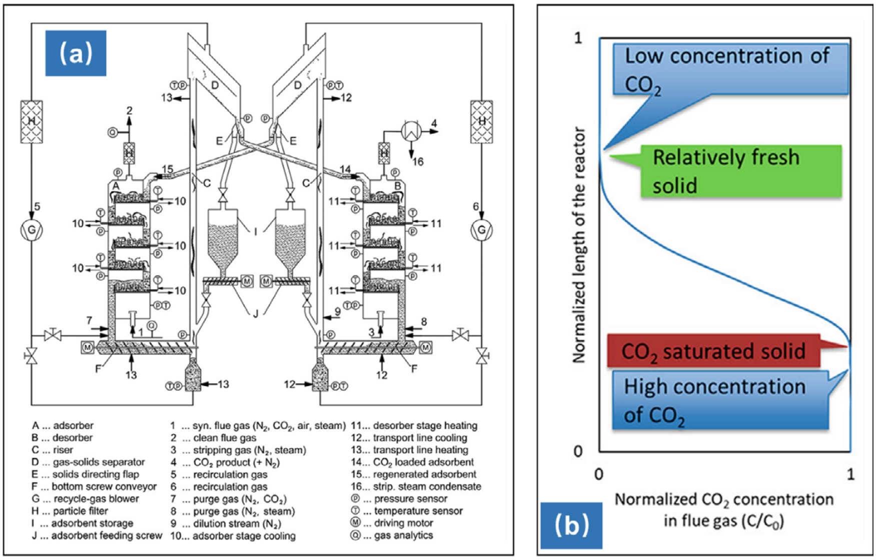


Fig. 7.1 CO_2 capture by temperature swing adsorption in a fluidized bed. **a** multi-stage fluidized bed (reprinted from ref. [8] with permission of Elsevier); **b** gas phase CO_2 concentration distribution along with the length of adsorber in (**a**) (from ref. [1], licensed under CC-BY 4.0)

regimes. In contrast to a conventional bubbling fluidized bed with a single regime, the gas velocity is decoupled in the bubbling and transport bed, thus facilitating the independent and flexible adjustment of the gas–solid contact time and sorbent circulation rate. The advantages of this reactor design were further evidenced by a 24-h test, which showed a low attrition rate and good cyclic stability of K_2CO_3/Al_2O_3 sorbent. As shown in Fig. 7.2b, further progress was made by the same group, researchers from which developed a two-stage integrated bubbling-transport reactor [14]. In comparison with the one-stage reactor shown in Fig. 7.2a, two-stage configurations can additionally introduce a quasi-countercurrent flow between CO_2 and K_2CO_3/Al_2O_3 through returning CO_2-lean sorbents to stage I. As a result, this quasi-countercurrent flow can largely promote the CO_2 capture efficiency.

The second important concern for low-temperature CO_2 sorption is the efficient heat management to reduce the energy requirement for the sorbent regeneration and the system operation. Concerning sorbent regeneration, low-grade heat like low pressure steam would normally be used, while the steam extraction would largely reduce power generation efficiency. More economic options for sorbent regeneration have been proposed, including integrating solar energy, collecting the waste heat released from the adsorption reactor, etc. Zaabout et al. [15, 16] proposed a concept of transient FBRs based on the swing adsorption reactor cluster (SARC, see Fig. 7.3) for low-temperature adsorption-based CO_2 capture, which alternately performs the carbonation, evacuation, regeneration and cooling steps without the need for sorbents recirculation. Moreover, this system employs a heat pump to manage heat between adsorption and regeneration reactors, thus reducing the energy requirement for sorbent regeneration. Another merit of this configuration is that the temperature difference between the adsorption and regeneration can be largely minimized by combining the vacuum and temperature swing adsorption (VTSA). Lab-scale of SARC indicates that the required temperature swing can be reduced by 30–40 °C at a 50 mbar vacuum [16], leading to a highly efficient heat transfer via the heat pump and significant energy saving for sorbent regeneration.

7.4 Scale-Up Demonstrations

To date, most research on low-temperature CO_2 sorption has been conducted at the lab scale, while merely a few works reported the demonstration at the pilot scale, as listed in Table 7.1. The Korea Institute of Energy Research (KIER) has made many endeavors to scale up the low-temperature CO_2 capture technology based on K_2CO_3 sorbents. In 2009, KIER first demonstrated the feasibility of the K_2CO_3-based low-temperature CO_2 sorption under real flue gas conditions from a 2 MW coal-fired plant. By employing a fast FBR for CO_2 adsorption at 70–90 °C and a bubbling FBR for regeneration at 160 °C, an average CO_2 capture efficiency of $\sim$ 70% was achieved [9]. Moving forward to 2014, KIER testified to the operation of a 10 MWe CO_2 capture system in Korea. This system was fed with 35,000 Nm^3/h flue gas from a 500 MW coal-fired power plant, and possessed the ability to capture ~ 200 tCO_2/

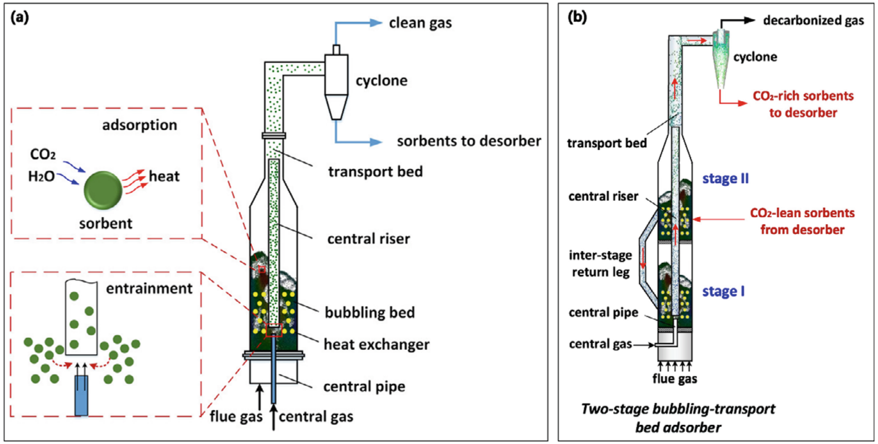


Fig. 7.2 **a** Integrated bubbling-transport FBR (reprinted from ref. [13] with permission of American Chemical Society); **b** two-stage integrated bubbling-transport FBR (reprinted from ref. [14] with permission of Elsevier)

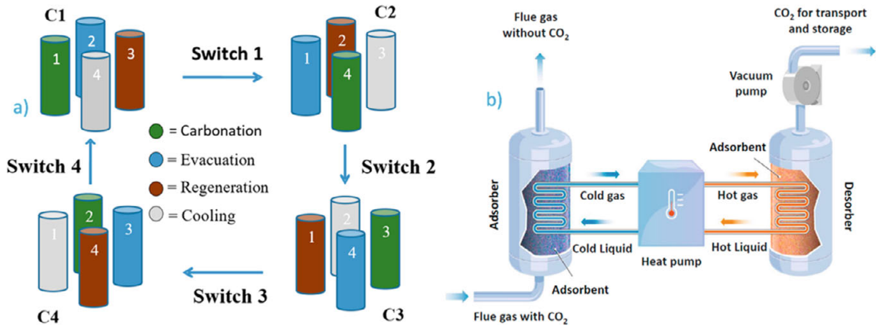


Fig. 7.3 Transient fluidized bed based on the SARC concept: **a** a cluster of fluidized reactors for continuous operation; **b** heat management in SARC by using a heat pump (reprinted from ref. [16] with permission of Elsevier)

d by using an inter-connected FBR, in which K_2CO_3-based sorbents were operated at 40–80 °C and 140–200 °C to capture and release CO_2, respectively. Simulation studies estimated the energy penalty of such a CO_2 capture system, indicating that 4–5 MJth/kg was required to reach a CO_2 capture efficiency of 80% and a CO_2 purity of 95% [17].

ADA Environmental Solutions (ADA) has been working on incorporating solid sorbents with fluidized bed technology for efficient low-temperature CO_2 capture from power plant flue gas. An amine-based ion exchange resin was screened out from more than 200 candidates. In 2014, they successfully constructed and executed a 1 MWe-scale process for low-temperature CO_2 capture installed at a coal-fired power plant in Alabama, USA [11]. Specifically, such 1 MWe configuration comprises a three-staged FBR for CO_2 capture at 40 °C, which is integrated with a single-staged fluidized bed regenerator operated at 120 °C. Based on their developed amine-based sorbents, ADA conducted a detailed techno-economic analysis. The reported capital cost of their configuration ranged from 124 \$/tCO_2 to 256 \$/tCO_2 at a CO_2 capture rate of 90%, which significantly outperformed a benchmark MEA system. Additionally, an environmental assessment was also performed on the amine-based sorbents selected for use. ADA testified to their sorbents as nonhazardous according to the OSHA Hazard Communication Standard [18].

RTI International applied its three-staged fluidized bed CO_2 capture process to flue gas from various industrial sources, including coal-fired power plants, NGCC power plants, and cement production plants. RTI claimed that technical and economic barriers of low-temperature CO_2 capture by solid sorbents could be overcome through the high-performing PEI/silica sorbents, operating at 60–70 °C for CO_2 adsorption and 110–120 °C CO_2 for release, respectively [10]. The estimated cost for RTI's process is around 39.7 \$/tCO_2, which is of high potential to meet the U.S. DOE's goal of > 90% CO_2 capture rate for 40 \$/tCO_2 captured by 2025.

7.5 Research Gaps and Future Trends

As previously mentioned, fluidized bed configurations are appropriate for the TSA-based process and have been verified by many institutions. However, there still exist many challenges in terms of practicality, scalability, and operation cost. When applying the TSA process in fluidization reactors, the employment of a lean/rich heat exchanger indicates a regular practice to minimize the energy penalty of the system. One important challenge lies in the design and integration of such a heat exchanger to perform the efficient heat deletion, from the adsorption reactor and the addition of the regeneration reactor, respectively. It has been reported that 20% or higher energy can be saved if an efficient heat exchanger is set up to collect the heat from the hot sorbents in the regenerate reactor and to preheat the cold sorbent in the adsorption reactor [19]. Existing solid–solid heat exchangers have significant drawbacks, such as bulky equipment, operation complexity, and poor heat-exchange efficiency. Further investigations should focus on heat integration via a heat pump and the incorporation of solar energy with FBRs. Several novel concepts have already been proposed on these aspects but remain far from industrial applications [16, 20, 21].

Another typical challenge for FBRs is that the well-mixed sorbents particle inherently results in early CO_2 breakthrough. As a result, CO_2 capture efficiency is impaired due to the low sorbent utilization rate. The multi-staged fluidized bed manifests a promising configuration to enhance gas–solid interactions, thus improving the CO_2 capture efficiency. As mentioned above, many pilot-scale demonstrations for low-temperature CO_2 capture have already been constructed with such promising configurations. Nonetheless, designing an FBR is still challenging and should be carefully studied in the future, not only to achieve a desirable heat integration, but also to maintain a good balance between the gas–solid mass transfer and the pressure drop. Though incremental stages in the fluidized bed could benefit energy savings by reducing the circulation rate, the additional pressure drop caused by the distributor plate in each stage should be noteworthy, which may significantly increase the energy penalty. Regarding this, Veneman [22] proposed a multistage based configuration characterized with diluted particle concentration in the adsorption reactor (void fraction > 90%). Such configuration allows the operation at high velocity with low pressure, thus greatly alleviating the pressure drop issue.

In addition, to achieve the counter-current mode in the multi-staged fluidized bed, each stage requires a downcomer to facilitate the downward particle transfer across the stages. These additional internal components in the multi-staged fluidized bed will result in higher complexity and operation costs, in contrast to a conventional single-staged fluidized bed. Hence, the multi-staged configuration will inherently confront with considerable scale-up challenges that should be carefully noted in the future, for instance, to investigate the thermodynamic system, model the hydrodynamics process, evaluate the risk of sorbent attrition, analyze the techno-economic feasibility, etc.

7.6 Ending Remarks

Low-temperature CO_2 sorption technology has gained increasing research focus on the incorporation of solid sorbents with FBRs. In this chapter, the process fundamentals of low-temperature CO_2 sorption were first introduced from the aspects of physisorption and chemisorption. The current progress of implementing fluidized bed configurations for low-temperature CO_2 capture was reviewed and highlighted, involving the process integration and the scale-up demonstrations. Nonetheless, this technology still has to overcome plenty of challenges for the industrial scale-up, for instance, to optimize the adsorption reactor configuration and reduce the regeneration penalty and operation cost. Further efforts in these areas are essential to advance the scale-up of low-temperature CO_2 capture technologies based on FBRs.

References

1. Dhoke C, Zaabout A, Cloete S, Amini S (2021) Review on reactor configurations for adsorption-based CO_2 capture. Ind Eng Chem Res 60:3779–3798
2. Yu CH, Huang CH, Tan CS (2012) A review of CO_2 capture by absorption and adsorption. Aerosol Air Qual Res 12:745–769
3. Gao W, Liang S, Wang R, Jiang Q, Zhang Y, Zheng Q et al (2020) Industrial carbon dioxide capture and utilization: state of the art and future challenges. Chem Soc Rev 49:8584–8686
4. Dhoke C, Zaabout A, Cloete S, Seo H, Park YK, Demoulin L et al (2020) Demonstration of the novel swing adsorption reactor cluster concept in a multistage fluidized bed with heat-transfer surfaces for postcombustion CO_2 capture. Ind Eng Chem Res 59:22281–22291
5. Ooi ZL, Tan PY, Tan LS, Yeap SP (2020) Amine-based solvent for CO_2 absorption and its impact on carbon steel corrosion: a perspective review. Chin J Chem Eng 28:1357–1367
6. Cai T, Chen X, Tang H, Zhou W, Wu Y, Zhao C (2021) Unraveling the disparity of CO_2 sorption on alkali carbonates under high humidity. J CO_2 Utilization 53:101737
7. Zhao C, Chen X, Zhao C (2010) Multiple-cycles behavior of K_2CO_3/Al_2O_3 for CO_2 capture in a fluidized-bed reactor. Energy Fuels 24:1009–1012
8. Schöny G, Dietrich F, Fuchs J, Pröll T, Hofbauer H (2017) A multi-stage fluidized bed system for continuous CO_2 capture by means of temperature swing adsorption-first results from bench scale experiments. Powder Technol 316:519–527
9. Park YC, Jo SH, Ryu CK, Yi CK (2009) Long-term operation of carbon dioxide capture system from a real coal-fired flue gas using dry regenerable potassium-based sorbents. Energy Procedia 1:1235–1239
10. Nelson TO, Kataria A, Mobley P, Soukri M, Tanthana J (2017) RTI's solid sorbent-based CO_2 capture process: technical and economic lessons learned for application in coal-fired, NGCC, and cement plants. Energy Procedia 114:2506–2524
11. Sjostrom S, Krutka H (2010) Evaluation of solid sorbents as a retrofit technology for CO_2 capture. Fuel 89:1298–1306
12. Schöny G, Infantino M, Fuchs J, Pröll T, Bertsch O, Höckner E et al (2017) The ViennaGreenCO_2 project-demonstration of low temperature solid sorbent CO_2 capture at pilot scale. In: 74th IEA-FBC meeting
13. Ma J, Xu Y, Wu Y, Chen X, Cai T, Liu D (2019) Continuous CO_2 capture performance of K_2CO_3/Al_2O_3 sorbents in a novel integrated bubbling-transport fluidized reactor. Ind Eng Chem Res 58:19733–19740
14. Ma J, Zhong J, Bao X, Chen X, Wu Y, Cai T et al (2021) Continuous CO_2 capture performance of K_2CO_3/Al2O3 sorbents in a novel two-stage integrated bubbling-transport fluidized reactor. Chem Eng J 404:126465
15. Zaabout A, Romano MC, Cloete S, Giuffrida A, Morud J, Chiesa P et al (2017) Thermodynamic assessment of the swing adsorption reactor cluster (SARC) concept for post-combustion CO_2 capture. Int J Greenhouse Gas Control 60:74–92
16. Dhoke C, Zaabout A, Cloete S, Seo H, Park Y-K, Blom R et al (2019) The swing adsorption reactor cluster (SARC) for post combustion CO_2 capture: experimental proof-of-principle. Chem Eng J 377:120145
17. Park YC, Jo SH, Kyung DH, Kim JY, Yi CK, Ryu CK et al (2014) Test operation results of the 10 MWe-scale dry-sorbent CO_2 capture process integrated with a real coal-fired power plant in Korea. In: 12th international conference on greenhouse gas control technologies, Ghgt-12
18. Ocker K (1988) OSHA's Hazard communication standard. Idaho Law Rev 619
19. Pirklbauer J, Schöny G, Pröll T, Hofbauer H (2018) Impact of stage configurations, lean-rich heat exchange and regeneration agents on the energy demand of a multistage fluidized bed TSA CO_2 capture process. Int J Greenhouse Gas Control 72:82–91
20. Bonaventura D, Chacartegui R, Valverde J, Becerra J, Verda V (2017) Carbon capture and utilization for sodium bicarbonate production assisted by solar thermal power. Energy Convers Manage 149:860–874

21. Tregambi C, Troiano M, Montagnaro F, Solimene R, Salatino P (2021) Fluidized beds for concentrated solar thermal technologies-a review. Front Energy Res 9:618421
22. Veneman R, Hilbers T, Brilman DWF, Kersten SRA (2016) CO_2 capture in a continuous gas-solid trickle flow reactor. Chem Eng J 289:191–202

Open Access This chapter is licensed under the terms of the Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/), which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license and indicate if changes were made.

The images or other third party material in this chapter are included in the chapter's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the chapter's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.

Chapter 8
Challenges and Outlooks

The utilization of FBR offers numerous advantages for implementing innovative CO_2 capture technologies. However, these novel CO_2 capture technologies introduce unique challenges due to their distinct properties compared to conventional applications.

8.1 Technical Hurdles and Economic Barriers

One principal challenge lies in the complexity of hydrodynamics within the reactors. This complexity is crucial because the fluidization quality directly influences the efficiency of gas–solid contact, which is critical for both oxy-fuel and chemical looping combustion processes. Moreover, the efficiency of FBRs is further challenged by the propensity for agglomeration and channeling, phenomena that can drastically reduce the capture efficiency of CO_2. Particularly in the calcium looping process, where the reactivity of lime diminishes with each carbonation/calcination cycle, there is a continuous need for constant replenishment of fresh sorbent and disposal of spent material. To address these multifaceted challenges, a multi-disciplinary approach is required, integrating advanced computational fluid dynamics modeling, innovative reactor design, and novel materials engineering. In reactor design, special attention must be given to the design of gas distributors and solids recycling systems to ensure uniform fluidization and minimize dead zones within the reactor. Additionally, the design of the fuel reactor in CLC systems must be optimized to handle the highly endothermic reduction of oxygen carriers, a step critical for ensuring adequate heat supply and maintaining reaction kinetics favorable for CO_2 capture.

Heat and material transport are critical factors across all the CO_2 capture technologies employing CFB. The circulation of solids between two reactors (CLC, CaL, and low-temperature CO_2 capture) and flue gas recirculation in oxy-fuel CFB has a significant influence on the temperature control, material balance, and dynamic

© The Author(s) 2025

L. Duan and D. Lu, *Fluidized Bed Reactors for Carbon Capture*,
https://doi.org/10.1007/978-981-96-0274-2_8

stability due to the feedback mechanisms involving heat and material. In CLC, the solids circulating rate impacts not only the temperature control, as the oxidation of fuel may be endothermic, but also the oxidation state of the oxygen carrier in the FR. The latter further affects fuel conversion rates and selectivity towards CO_2, ultimately influencing CO_2 capture efficiency. Additionally, strategies for maintaining partial load stability of the FBRs will have to be investigated, as peak shaving is often required. The Recycled flue gas (RFG) in oxy-fuel combustion offers operational flexibility and may be beneficial for CLC. Similarly, in CaL, a more adaptable carbonator, such as a bubbling-turbulent fluidized bed, could be an option to facilitate load adaptation.

Transitioning FBR technology from the pilot scale to full industrial deployment is an enormous challenge. It involves scaling up the reactor size by orders of magnitude while maintaining the same efficiency and capture rates achieved in smaller-scale models. This challenge is complex as the scale-up is not linear and requires a detailed understanding of the scaling laws that govern fluidization, reaction kinetics, and heat and mass transfer. Moreover, addressing reactive issues, such as degradation, attrition, and the match-up of different reaction rates between carbonation/calcination in the CaL process, oxidation/reduction in the CLC process, as well as sorption/desorption in the low-temperature sorption process, is imperative prior to the scale-up of FBRs.

Establishing a reliable and generalized criterion is crucial for the scale-up of the FBR. Specifically, the scale-up of FBR traditionally relies on the hydrodynamic similarity criteria, which are based on dimensionless analysis according to the Buckingham π-theorem [1]. However, it is important to note that similar hydrodynamic behaviors do not always guarantee similar reactor performance, e.g., fuel conversion and CO_2 capture efficiency. Thus, a more comprehensive approach is needed. Strategies for industrial scale-up must be rooted in rigorous experimentation and modeling. This approach should involve pilot programs that progressively increase in scale, as these programs can provide valuable data to inform the design of commercial-scale reactors. Moreover, partnerships with industry can facilitate the sharing of knowledge and resources, which is essential for accelerating the development and adoption of FBRs in the marketplace.

Besides, it is significant to develop modeling strategies that bridge the gaps between different scales involved in the FBR, in order to perform simulations at an industrial-relevant scale under typical operating conditions in a reasonable calculating time. The gap-bridging strategies include, for instance, the widely used Langmuir–Hinshelwood-Hougen-Watson type rate equation, which is concise and represents the detailed reaction mechanism with certain elementary steps being the rate-determining step [2]. The intra-particle and gas-particle heat and mass transfer are well-documented, and consensus is generally achieved. Recently, advanced techniques have been used to study the validity of the classical constitutive equations. For example, the CFD simulation and the infra-red technique were employed to investigate the bubble-emulsion mass transfer coefficient, while X-ray tomography and electrical capacitance tomography were used to analyze the bubbling behavior [3, 4]. These studies have reinforced the accuracy of the two-phase model. Concerning the solids mixing, CFD and radiotracer techniques were adopted to research the solids mixing and residence time distribution [5, 6]. Therefore, it is possible to develop a model that accounts for the essential phenomena in the FBR while keeping the calculating time reasonable for industrial-scale reactors [7].

Understanding the economic feasibility of FBRs for carbon capture is a complex issue. Initially, significant capital expenditure is required, mainly due to the costs associated with constructing large-scale reactors, procuring oxygen carriers and sorbents, and integrating these systems with existing power generation facilities. Moreover, the operational costs, which include energy penalties incurred from the increased fuel requirements to meet the thermal demands of the capture process, presenting significant economic challenges. To address these financial concerns, innovation in material sciences is crucial to extend the lifespan of sorbents and oxygen carriers. Similarly, advancements in reactor design aimed at minimizing energy consumption are essential.

What's more, it is vital to conduct a thorough techno-economic assessment that integrates life cycle analysis with real-world operational data. This assessment is necessary to identify cost reduction opportunities and to compare the economic viability of FBRs with other CO_2 capture technologies. Such evaluations will provide a more comprehensive understanding of the cost–benefit landscape, guiding future developments and implementations of FBR technology.

8.2 Innovation and Future Research Priorities

The successful implementation of FBRs in carbon capture critically depends on the development of materials that can endure the stringent conditions of the process. It is essential that oxygen carriers and sorbents are not only robust but also possess high reactivity and resistance to sintering, attrition, and agglomeration. Efficiently integrating these materials into a process that maximizes efficiency and minimizes waste adds another dimension of complexity. Therefore, innovation in materials requires a collaborative and interdisciplinary approach, encompassing fields such as material science, chemistry, and engineering. Engaging in collaboration with industry partners is key to bridging the gap between laboratory-scale research and the practical requirements of real-world applications. Moreover, the exploration of advanced manufacturing techniques, like 3D printing, could open up new avenues for producing complex reactor components with superior performance characteristics.

In shaping the future of FBRs in carbon capture, collaborative research initiatives that unite experts across various disciplines will play a pivotal role. These collaborations have the potential to significantly accelerate the pace of innovation, spanning from fundamental research to the development of pilot projects, and ultimately, to commercial-scale operations. Creating an ecosystem that promotes open communication and resource sharing is fundamental to overcoming the technical, economic, and regulatory challenges currently impeding progress. Such collaborative initiatives should strive to establish consortia that leverage the strengths of each participant, including academic institutions with advanced research capabilities, industry partners with practical experience and resources, or government bodies capable of influencing policy and providing funding. Through these synergistic partnerships, the field can advance collectively, positioning FBRs as a feasible and effective solution for carbon capture in the global effort to combat climate change.

The economic perspective for carbon capture is changing, with increasing recognition of the need to price carbon emissions accurately. Economic incentives, such as tax credits for Carbon Capture, Utilization, and Storage (CCUS) and tradeable carbon allowances, have the potential to substantially enhance the viability of FBRs in this context. By integrating the cost of carbon emissions into the economic framework, these mechanisms can tip the balance in favor of carbon capture technologies.

However, the development of a robust CCUS strategy is not a solitary endeavor but requires a collaborative approach involving governments, industry, and academia. Such a collaborative effort is crucial to ensure that the economic incentives are designed effectively, not only promoting the adoption and development of FBRs but also fostering a market for captured CO_2. This market could encompass its utilization in industrial processes or its permanent sequestration. Through these combined efforts, a sustainable and economically feasible framework for carbon capture can be established, aligning environmental objectives with economic incentives.

References

1. Yates JG, Lettieri P (2016) Fluidized-bed reactors: processes and operating conditions. Springer, Switzerland
2. Froment GF, Bischoff KB, De Wilde J (1990) Chemical reactor analysis and design. Wiley, New York
3. Hernández-Jiménez F, Gómez-García A, Santana D, Acosta-Iborra A (2013) Gas interchange between bubble and emulsion phases in a 2D fluidized bed as revealed by two-fluid model simulations. Chem Eng J 215:479–490
4. Medrano J, Gallucci F, Boccia F, Alfano N, van Sint AM (2017) Determination of the bubble-to-emulsion phase mass transfer coefficient in gas-solid fluidized beds using a non-invasive infra-red technique. Chem Eng J 325:404–414
5. Luo K, Wu F, Yang S, Fang M, Fan J (2015) High-fidelity simulation of the 3D full-loop gas-solid flow characteristics in the circulating fluidized bed. Chem Eng Sci 123:22–38
6. Pant H, Sharma V, Goswami S, Samantray J, Mohan I, Naidu T (2014) Residence time distribution study in a pilot-scale gas-solid fluidized bed reactor using radiotracer technique. J Radioanal Nucl Chem 302:1283–1288
7. He Z, De Wilde J (2021) Numerical simulation of commercial scale autothermal chemical looping reforming and bi-reforming for syngas production. Chem Eng J 417:128088

Open Access This chapter is licensed under the terms of the Creative Commons Attribution 4.0 International License (http://creativecommons.org/licenses/by/4.0/), which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license and indicate if changes were made.

The images or other third party material in this chapter are included in the chapter's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the chapter's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder.