

Contents lists available at ScienceDirect

Chemical Engineering Journal



journal homepage: www.elsevier.com/locate/cej

Study of biomass gasification combined with CO₂ absorption in a dual fluidized bed (DFB) using the Eulerian-Lagrangian method



Mengyao Guo^a, Jiahui Yu^a, Shuai Wang^{a,b}, Kun Luo^{a,b,*}, Jianren Fan^{a,b}

^a State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou 310027, China
 ^b Shanghai Institute for Advanced Study of Zhejiang University, Shanghai 200120, China

ARTICLE INFO

Keywords: Dual fluidized bed Biomass gasification CO₂ absorption MP-PIC Numerical simulation

ABSTRACT

Biomass absorption enhanced reforming (AER) gasification technology uses CO2 absorbent as bed material, which can cleanly and efficiently realize the enrichment of H₂ and the removal of CO₂ in the product gas. However, the in-depth understanding of complex gas-solid flow characteristics and thermochemical behavior in the AER gasification process is still lacking. This work integrates heat transfer and complex reaction kinetics regarding gasification, carbonation, and calcination based on the multiphase particle-in-cell (MP-PIC) method. After model validation, biomass gasification in a pilot-scale dual fluidized bed (DFB) system is numerically studied under the conventional gasification and AER gasification conditions. The physical-thermo-chemical behavior under two gasification conditions is quantitatively compared and analyzed, and the influence of several key operating parameters on gasification performance is investigated under the AER gasification condition. The results show that compared with conventional gasification, AER gasification has the characteristics of (i) the particle movement is more gentle, which is beneficial to the heat and mass transfer process between the bed material and the biomass particles; (ii) the temperature difference in the two reactors is larger and the temperature of reactors is lower, which is beneficial to the carbonation in the gasifier; (iii) it reduces the mole fraction of CO_2 by 76.67 %, increases the mole fraction of H_2 by 54.58 %, which greatly improves the quality of the gas products. In addition, under the AER gasification condition, increasing the gasification temperature promotes the carbonation reaction and methane steam reforming reaction, thereby improving the gasification performance. The change in steam flow rate has no significant impact on the gasification performance. Furthermore, properly adjusting the content of absorbent can reduce the cost of bed materials. The present work provides a cost-effective approach to gaining insight into the physical-thermo-chemical behavior of AER gasification.

1. Introduction

As global warming intensifies and the energy crisis worsens, biomass has gathered lots of attention from researchers as an environmentally friendly renewable energy source [1–4]. Compared to traditional fossil fuels, biomass resources are abundant and low-cost. Moreover, the amount of CO₂ accumulated by biomass during its growth is equal to the amount of CO₂ released, enabling carbon neutrality throughout its lifecycle [1,5]. The stored energy in biomass can be utilized through both combustion and gasification processes. However, direct biomass combustion will produce a significant number of pollutants and pollute the environment. In contrast, biomass gasification not only generates heat and electricity but also yields valuable gases, such as CO and H₂ (syngas). Therefore, biomass gasification technology currently represents one of the predominant approaches for biomass energy utilization [6,7].

Biomass gasification for hydrogen production refers to the thermochemical conversion of biomass into syngas. This process is carried out under high-temperature conditions using gasification agents, such as air, oxygen, steam, and carbon dioxide. The choice of gasification agent has a significant influence on the volume fraction of hydrogen and the heating value of the syngas [8]. The addition of steam during the gasification process will enhance various reaction processes, including gasification reaction, water–gas shift reaction, and methane steam reforming reaction, leading to increased hydrogen generation. Besides, biomass steam gasification exhibits three times higher hydrogen production rates compared to air gasification, with lower by-product

* Corresponding author at: State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou 310027, China. *E-mail address:* zjulk@zju.edu.cn (K. Luo).

https://doi.org/10.1016/j.cej.2024.148723 Received 17 October 2023; Received in revised form 3 January 2024; Accepted 10 January 2024 Available online 13 January 2024

1385-8947/© 2024 Elsevier B.V. All rights reserved.

Nomenc	lature	$Y_{g,i}$	Mass fraction of i^{th} gas specie (–)
$\begin{array}{c} A\\ A_p\\ C_p\\ C_V\\ d_p\\ D_{g,i}\\ D_p\\ f_D\\ F_{gp}\\ \mathbf{g}\\ h_g\\ \Delta H_{rg}\\ k\end{array}$	Particle acceleration (m/s^2) Particle surface area (m^2) Specific heat capacity of particle $(J/(kg\cdot K))$ Specific heat capacity of gas phase $(J/(kg\cdot K))$ Diameter of the solid phase (m) Mass diffusion coefficient of gas (m^2/s) Drag function coefficient $(-)$ Distribution function of particle $(-)$ Interphase force between the gas and particle phases (N) Gravitational acceleration (m/s^2) Specific enthalpy of gas (J/kg) Heat source forming from chemical reactions (W/m^3) Papation rate coefficient $(I/(m-s))$	Greek syn γ, ξ θ θ_{cp} ε_p λ_g μ_g ρ σ τ_D τ_g τ_p	nbols Model parameter (-) Volume fraction (-) Solid volume fraction at close-packing state (-) Particle emissivity (-) The molecular conductivity of the gas phase (W/(m·K)) Gas shear viscosity (kg/(m·s)) Density (kg/m ³) Stefan-Boltzmann constant W/(m ² ·K ⁴) Particle collision damping time (s) Gas stress tensor (Pa) Inter-particle stress (Pa)
k m MW _{p,i} δṁ _p δṁ _{i,r}	Reaction rate coefficient (L/(g _{cat} ·s)) Mass (kg) molar mass of particle <i>i</i> Mass source term (kg/(m ³ ·s)) The net consumption or production rate of gas specie <i>i</i> (kg/ (m ³ ·s))	Subscript: g i P Acronym	s Gas phase Particle <i>i</i> Particle phase
Nu P_g P_p P_{CO2} Pr q \dot{Q}_D Re S_{gp} S_{gw} t T \boldsymbol{u}	Nusselt number (-) Pressure of gas phase (Pa) Pressure constant (Pa) Partial pressure of CO ₂ (Pa) Prandtl number (-) Heat flux (W/m ²) Enthalpy diffusion (W/m ³) Reynolds number (-) Gas-solid energy exchange rate (W/m ³) Gas-wall convective heat transfer (W/m ³) Time instant (s) Temperature (K) Velocity(m/s)	AER BFB CFD CFD-DEN CGC DFB LHV MP-PIC PDF PSD RME	Absorption enhanced reforming Bubbling fluidized bed Circulating fluidized bed Computational fluid dynamics Computational fluid dynamics – discrete element method Combustible gas (e.g., H ₂ , CO, CH ₄) concentration Dual fluidized bed Lower heating value Multiphase particle-in-cell Particle distribution function Particle size distribution Rapeseed methyl ester

content, thereby making it the first choice for hydrogen production from biomass gasification [9]. However, one major drawback of biomass gasification is the presence of CO_2 in the syngas, resulting in a significant reduction in the calorific value and usable value of syngas. Consequently, the syngas usually cannot be directly used. To address this issue, some researchers have proposed the addition of solid CO_2 adsorbents during the gasification process to decrease CO_2 in the product gas and simultaneously increase the concentration of combustible gases, primarily hydrogen. This process is known as absorption enhanced reforming (AER) gasification. Calcium-based limestone, which possesses high carbon dioxide absorption capacity, catalytic activity, and costeffectiveness, is commonly employed as the bed material for hydrogen-rich gasification [10–12].

Fluidized bed gasification systems can achieve improved gas-solid mixing efficiency and reaction rates, making it a promising option for large-scale commercialization [6]. According to the heat transfer modes, fluidized bed gasifiers can be divided into auto-thermal gasifiers and hetero-thermal gasifiers [13]. The auto-thermal fluidized bed gasifiers supply heat directly by the oxidation reaction within the gasifier, typically using air or pure oxygen as the gasification agent. However, this approach reduces the heating value of product gas or requires expensive air separation units [14]. Conversely, the hetero-thermal gasifier employs an external heat source for providing heat, commonly utilizing steam as the gasification agent. The dual fluidized bed (DFB) is



Fig. 1. Schematic diagram of biomass AER gasification process [4].

Table 1

Chemical reaction and reaction rates [43,44].

Chemical reaction equation	Chemical reaction rate
R4: C + $O_2 \rightarrow CO_2$	$r_4 = 4.34 \times 10^7 m_c T_p \exp(-13590/T_p)[O_2]$
$\text{R5: } C + H_2 O \mathop{\leftrightarrow} CO + H_2$	$r_{5f} = 6.36m_c T_p \exp(-22645/T_p)[H_2O]$
	$r_{5r} = 5.218 \times 10^{-4} m_c T_p^2 \exp(-6319/T_p - 17.29)$
	$[H_2][CO]$
R6: C + CO ₂ \leftrightarrow 2CO	$r_{6f} = 6.36m_c T_p \exp(-22645/T_p)[CO_2]$
	$r_{6r}=5.218\times$
	$10^{-4}m_{\rm c}T_{\rm p}^2 {\rm exp}(-2363/T_{\rm p}$ - 20.92)[CO] ²
$\text{R7: } C + 2H_2 \mathop{\leftrightarrow} CH_4$	$r_{7f}~=6.838\times$
	$10^{-3}m_{\rm c}T_{\rm p}\exp(-8078/T_{\rm p}-7.087)[{\rm H_2}]$
	$r_{7r}=0.755 m_c T_p^{0.5} exp(-13578/T_p0.372) [\text{CH}_4]^{0.5}$
R8: $CH_4 + H_2O \rightarrow CO + 3H_2$	$r_8 = 3 \times 10^5 exp(\ \text{-}\ 15042/T_g) [\text{CH}_4] [\text{H}_2\text{O}]$
R9: CO + H ₂ O \rightarrow CO ₂ + H ₂	$r_9~=7.68\times 10^{10} exp(-36640/T_g) [CO]^{0.5} [H_2O]$
R10: $C_{16}H_{34}$ +	$r_{10}=3.8\times10^{11} exp($ - $1.255\times10^8/$
$24.5O_2 \rightarrow 16CO_2 + 17H_2O$	$RT_{\rm g})[{\rm C}_{16}{\rm H}_{34}]^{0.25}[{\rm O}_2]^{1.5}$



Fig. 2. The equilibrium partial pressure of CO_2 ($P_{CO2, eq}$) as a function of temperature [45,46].

considered a typical hetero-thermal gasification system, which offers noteworthy advantages such as high efficiency, intensified gasification, and exceptional fuel adaptability and has gained widespread usage globally. A typical DFB system primarily comprises two independent reactors: a gasifier and a combustor [14]. Within the gasifier, biomass fuel is converted into syngas with a high heating value through a series of processes including drying, pyrolysis, and gasification. The bed material flows into the combustor along with the remaining char particles, where the remaining char and supplementary fuel combust to heat the bed material and provide the heat for the gasification reaction in the gasifier.

Fig. 1 shows the schematic diagram of the biomass AER gasification process. The main component of the circulating bed material is usually CaO/CaCO₃, which can not only act as the heat carrier but also be used to selectively transport carbon dioxide in the product gas. CO₂ is absorbed in the gasifier and then released after being transported to the combustor, thereby reducing the CO₂ in the product gas and increasing the content of H₂. Due to the existence of carbonation reaction, the temperature in the gasifier is generally 600–700 °C under the AER gasification condition. The temperature of the calcination reaction in the combustor is 830–930 °C [15]. The reaction equation can be given by:

$$CaCO_3 \leftrightarrows CaO + CO_2$$
 (R1)

In the past few years, researchers have carried out a large number of experimental studies on the DFB biomass AER gasification system [16–18]. However, due to the difficulty of measurement under high-temperature conditions and the complexity of gasification reaction kinetics, the experimental research is time-consuming and difficult to carry out. At present, the comprehensive understanding of hydrody-namics and thermochemical characteristics during the biomass AER gasification process in the DFB system is still lacking. With the improvement of computer capacity and numerical algorithms, the utilization of computational fluid dynamics (CFD) for numerically investigating the process of fluidized bed biomass gasification has become an

Table 2				
Operating temperatures	under t	he two	gasification	condition

Reactor	Conventional gasification	AER gasification
Gasifier	838 °C	600 ~ 750 °C
Combustor	900 °C	894 °C



Fig. 3. Schematic diagram (a) [50,51], geometric model (b), and grids (c) of DFB.

Ultimate analysis (%)						Proximat	e analysis (%)			
С	Н	0	Ν	S	Cl	М	А	V	FC	LHV (KJ/Nm ³)
47.16	5.67	40.73	0.05	0.005	0.003	6.11	0.27	81.17	12.72	17.46

Table 4

Composition of the olivine.

Composition	MgO	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	CaO	NiO
Proportion(%)	46.2-48.1	37.6–40.4	7.7–10.1	0.8	<0.4	3.7

accurate, cost-effective, and reliable method. Alobaid et al. [19] provided a comprehensive review of multi-scale numerical approaches for simulating gas-solid flow in fluidized bed reactors, presenting detailed information on the development of 3D CFD models and their applications in various gas-solid fluidized bed systems. They also offered suggestions for further research in the field of 3D CFD simulation of fluidized beds. Moreover, CFD can provide crucial guidelines for the design, optimization, and expansion of reactor parameters [20,21]. Based on the treatment method of solid phase, existing multiphase numerical simulation methods for dense gas-solid two-phase flow can be divided into the Eulerian-Eulerian framework and the Eulerian-Lagrangian framework. The former embraces the continuum assumption and regards both the gas phase and the solid phase as continuous phases. Although this approach shows great computational efficiency, it falls short in accurately modeling gas-solid flow within the dilute phase region of the fluidized bed. Additionally, it fails to predict detailed particle-scale information, thereby limiting the study of microscopic

Table 5

Composition of the calcined calcite.

scale in fluidized equipment [22]. In contrast, under the Eulerian-
Lagrangian framework, the solid phase motion is solved under the
Lagrangian framework and detailed information about particles can be
obtained, such as particle trajectory and particle size change. Compu-
tational fluid dynamics - discrete element method (CFD-DEM) and
multiphase particle-in-cell (MP-PIC) are two typical methods under the
Eulerian-Lagrangian framework. The CFD-DEM method accurately
tracks individual particles and describes particle-particle collisions
using the soft sphere model. This approach enables precise resolution of
particle-scale information but requires a significantly reduced time step
and considerable computational resources. Consequently, its application
remains most appropriate for laboratory-scale calculations involving a
limited quantity of particles [22,23]. Compared with the CFD-DEM
method, the MP-PIC method introduces the concept of calculation par-
ticles and significantly reduces the number of calculation particles by
assigning one computation particle to represent multiple real particles
that have the same properties. Furthermore, the MP-PIC method sim-
plifies inter-particle collisions with a solid stress model, thereby further
reducing the computational requirements. These strategies enable the
MP-PIC method to capture particle-scale information also efficiently
calculate large-scale systems, balancing numerical accuracy and
computational efficiency [24,25]. Therefore, this method proves highly
suitable for simulating dense gas-solid reaction systems at both pilot and
industrial scales.

Composition	CaO	CaCO ₃	MgO	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	Trace elements
Proportion (%)	53.05	40.95	0.7	2.77	0.6	1.4	0.53



Fig. 4. Particle size distribution.

Table 6

Operating parameters of the DFB under the two gasification conditions.

Parameters	Conventional gasification	AER gasification
Gasifier		
Biomass feed rate (kg/s)	0.00695	0.00695
Biomass temperature (K)	298	298
Inlet steam flow rate (kg/s)	0.00253	0.00439
Steam temperature (K)	573	573
Combustor		
Primary air flow rate (kg/s)	0.00115	0.00058
Primary air temperature (K)	673	673
Secondary air flow rate (kg/s)	0.00706	0.00865
Secondary air temperature (K)	673	673
Gas products flow rate (Nm ³ /s)	0.046	0.035
Gas products temperature (K)	353	353
RME flow rate (kg/s)	0.00062	0.000225
RME temperature (K)	353	353
Operating pressure (MPa)	0.1	0.1
Solid volume fraction at close pack (-)	0.6	
Particle normal-to-wall retention coefficient (-)	0.9	
Particle tangential-to-wall retention coefficient (–)	0.3	
Fraction coefficient (-)	0.3	
Time step (s)	1.0×10^{-4}	

Table 7

Operating parameters of different cases under the AER gasification condition.

Case	T_b (°C)	Q_S (kg/s)	α (-)
Base case	650	0.00439	100 %
Gasification temperature	600	0.00439	100 %
	650	0.00439	100 %
	700	0.00439	100 %
	750	0.00439	100 %
Steam flow rate	650	0.00329	100 %
	650	0.00439	100 %
	650	0.00549	100 %
	650	0.00658	100 %
Absorbent content	650	0.00439	25 %
	650	0.00439	50 %
	650	0.00439	75 %
	650	0.00439	100~%

Over the past decade, the MP-PIC method has been successfully used in numerical studies of biomass gasification in fluidized bed systems. For instance, Liu et al. [26] studied the 1 WMth pilot-scale dual fluidized bed biomass gasification system. They found that the solids volume fraction in the lower regions of the combustor was significantly affected by particle size distribution (PSD), while the higher regions were not affected much. Sun et al. [27] conducted a simulation study on biomass gasification in an 8 WMth industrial-scale dual fluidized bed. The results showed that increasing the height of the solids stockpiles enhanced heat transfer performance, which lead to an increase of the vertical gas temperature in the gasifier while a decrease in the vertical gas temperature in the burner. Kong et al. [4] further studied the 8 MWth dual fluidized bed biomass AER gasification process. The results showed that the gas quality of AER gasification was greatly improved compared with the conventional gasification process. For the AER gasification process, increasing the gasification temperature and steam-to-biomass ratio improved the gasification performance. However, it should be pointed out that the present simulation works still focus on the conventional gasification process in the fluidized bed reactor, and mainly study the macroscopic motion of gas and solid phases. Few studies have been conducted on the biomass gasification process combined with the CO₂ absorption enhancement process.

To address the research gap, the present work introduces two key contributions. Firstly, we have developed a rigorously validated MP-PIC

reactive model that incorporates comprehensive sub-models, including gas turbulence, inter-particle/phase interactions, heat transfer, mass transfer, particle shrinkage, and complex reaction kinetics (e.g., gasification, carbonation, and calcination). Secondly, we have investigated the thermophysical characteristics of dense gas-solid reactive flow in a 100 KWth pilot-scale DFB system to uncover the underlying mechanism of biomass gasification. By numerically studying the effects of various key operating parameters on the gasification performance, we have demonstrated the superiority of AER gasification over conventional gasification. These findings provide valuable insights for reactor design and process optimization. The structure of this work is as follows: Section 2 details the gas-solid two-phase governing equations and the chemical reaction model. Section 3 presents the numerical setup and model validation. Section 4.1 analyzes the gasification process of conventional gasification and AER gasification from the aspects of gas-solid flow, temperature distribution, and gasification products. Section 4.2 discusses the influence of several key parameters on the performance of DFB biomass AER gasification. Finally, the conclusion is drawn in Section 5.

2. Numerical model

In the MP-PIC method, the gas motion is described by volumeaveraged Navier-Stokes equations, and the gas turbulence is solved by the method of large eddy simulation. The dynamics of solid phases are described by the particle distribution function (PDF), and the interparticle collision is simplified by introducing a solid stress model. Furthermore, detailed reaction kinetics including homogeneous and heterogeneous reactions are considered. The mathematical model is detailed as follows.

2.1. MP-PIC framework

For the gas phase, the conservation equations of mass, momentum, species, and energy are respectively given by [28,29]:

$$\frac{\partial(\theta_g \rho_g)}{\partial t} + \nabla \cdot (\theta_g \rho_g \boldsymbol{u}_g) = \delta \dot{m}_p \tag{1}$$

$$\frac{\partial \left(\theta_{g}\rho_{g}\boldsymbol{u}_{g}\right)}{\partial t} + \nabla \cdot \left(\theta_{g}\rho_{g}\boldsymbol{u}_{g}\boldsymbol{u}_{g}\right) = -\nabla \rho_{g} + \rho_{g}\theta_{g}\mathbf{g} + \nabla \cdot \left(\theta_{g}\boldsymbol{\tau}_{g}\right) + \boldsymbol{F}_{gp}$$
(2)



Fig. 5. Axial distribution of time-averaged temperature along the gasifier's centerline under different grid resolutions.



Fig. 6. Time evolution of gas products mass fractions: (a) conventional gasification; (b) AER gasification.



Fig. 7. Comparison of simulation results and experimental data of time-averaged mole fractions: (a) conventional gasification; (b) AER gasification.

$$\frac{\partial(\theta_g \rho_g Y_{g,i})}{\partial t} + \nabla \cdot (\theta_g \rho_g \boldsymbol{u}_g Y_{g,i}) = \nabla \cdot (\theta_g \rho_g D_{g,i} \nabla Y_{g,i}) + \delta \dot{m}_{i,r}$$
(3)

$$\frac{\partial(\theta_{g}\rho_{g}h_{g})}{\partial t} + \nabla \cdot (\theta_{g}\rho_{g}\boldsymbol{u}_{g}h_{g}) = \theta_{g}\left(\frac{\partial p_{g}}{\partial t} + \boldsymbol{u}_{g}\cdot\nabla p_{g}\right) - \nabla \cdot (\theta_{g}q) + \dot{Q}_{D} + S_{gp} + S_{gw} - \Delta H_{rg}$$
(4)

where ρ_g , u_g , and p_g are the density, velocity, and pressure of the gas phase, respectively. θ_g and **g** are the voidage and gravitational acceleration. F_{gp} is the inter-phase momentum exchange term. $\delta \dot{m}_p$ is the source term that links the reaction of the discrete phase and continuous phase. τ_g is the stress tensor of the gas phase. $Y_{g,i}$ and $D_{g,i}$ are the mass fraction and diffusion coefficient of the gas phase *i*, respectively. $\delta \dot{m}_{i,r}$ is the quality changes caused by chemical reactions involved in phase *i*. h_g and \dot{Q}_D are the gas enthalpy and enthalpy diffusion. S_{gp} and S_{gw} are the gas–solid and gas-wall heat transfer. q is the fluid heat flux and ΔH_{rg} is the reaction heat. equation, which is a function of particle's spatial position, velocity, mass, temperature, and instant time. The equation is written as [30]:

$$\frac{\partial f}{\partial t} + \frac{\partial (f \boldsymbol{u}_p)}{\partial t} + \frac{\partial (f \mathbf{A})}{\partial \boldsymbol{u}_p} = \frac{f_D - f}{\tau_D} + \frac{f_G - f}{\tau_G}$$
(5)

where u_p is the particle velocity. f_D and τ_D are the particle distribution function at local equilibrium and the particle collision relaxation time, respectively. After the particle collision, the velocity tends to be the isotropic Gaussian distribution. f_G and τ_G are the relaxation time and particle collision functions in this state. **A** is the particle acceleration and is defined as [31]:

$$\mathbf{A} = \frac{d\boldsymbol{u}_p}{dt} = D_p \left(\boldsymbol{u}_g - \boldsymbol{u}_p \right) - \frac{\nabla p}{\rho_p} - \frac{\nabla \tau_p}{\rho_p \varepsilon_p} + \mathbf{g} + \frac{\overline{\boldsymbol{u}}_p - \boldsymbol{u}_p}{2\tau_D}$$
(6)

$$\tau_p = \frac{P_p \theta_p^{\nu}}{\max\left[\left(\theta_{cp} - \theta_p\right), \xi\left(1 - \theta_p\right)\right]} \tag{7}$$

$$\theta_p = \iiint f \frac{m_p}{\rho_p} dm_p d\boldsymbol{u}_p dT_p \tag{8}$$

The dynamics of the particle phase are described by solving the PDF



Fig. 8. Comparison of simulation results and experimental data of time-average temperature in combustor and gasifier: (a) conventional gasification; (b) AER gasification.

where \overline{u}_p is the local mass-averaged particle velocity. D_p is the momentum exchange coefficient between the gas–solid phase, namely the drag coefficient. τ_p is the solid normal stress. P_p , γ , ξ are constants, which respectively represent particle dynamics solid-phase pressure, particle concentration index, and non-singular number. θ_{cp} is the solid volume fraction at the close-packing state.

Drag force plays a crucial role in predicting gas-solid two-phase flow by characterizing the momentum exchange between gas and solid in the flow field. Therefore, accurately modeling drag force is of great importance for understanding the behavior of such systems. In DFB systems, the bed material can be classified as Geldart B particles, thus the WenYu-Ergun drag model is adopted to accurately describe the drag force [32]. The WenYu-Ergun model is based on the assumption of homogeneous conditions inside a control volume, which is a combination of the WenYu drag model and the drag model based on the Ergun equation. Despite the discontinuity of the model at 0.8, it is commonly used in the simulation of dense gas-solid flow in fluidized beds, and satisfactory results were obtained to demonstrate its reasonability [33–35]. The mass conservation equation for the particle phase can be expressed as:

$$\frac{dm_{p,i}}{dt} = \frac{\theta_s M W_{p,i}}{\rho_p \theta_p} m_p \frac{dC_{p,i}}{dt}$$
(9)

$$\frac{dm_p}{dt} = \sum_{i=1}^{N} \frac{dm_{p,i}}{dt}$$
(10)

where $MW_{p,i}$ and $C_{p,i}$ are the molar mass and specific heat capacity of particle *i*.

The energy conservation equation for the solid phase can be expressed as follows:

$$m_p C_p \frac{dI_p}{dt} = Q_{gp} + Q_{rad} - \Delta H_{react}$$
(11)

$$Q_{gp} = \frac{\lambda_g \mathrm{Nu}_p}{d_p} A_p \left(T_g - T_p \right)$$
(12)

$$Q_{rad} = \sigma \varepsilon_p A_p \left(T_{b,local}^4 - T_p^4 \right)$$
⁽¹³⁾

$$Nu_p = 0.6Re_p^{1/2}Pr^{1/3} + 2.0$$
(14)

$$\operatorname{Re}_{p} = \frac{\rho_{g} \theta_{g} |\boldsymbol{u}_{g} - \boldsymbol{u}_{p}| d_{p}}{\mu_{g}}$$
(15)

$$Pr = \mu_g C_{p,g} / \lambda_g \tag{16}$$

where particle Nusselt number Nu_p is calculated by the correlation proposed by Ranz and Marshall [36], which combines the particle Reynolds number Re_p and Prandtl number Pr. u_p and d_p are the velocity and diameter of the solid phase. μ_g is the gas viscosity. $C_{p,g}$ is the gas heat capacity. λ_g is the thermal conductivity of the gas. T_g and T_p are the temperature of gas and particles, respectively. $T_{b,local}$ is the temperature of the surrounding environment. A_p , σ , and ε_p represent the particle surface area, Stefan-Boltzmann constant, and particle emissivity, respectively.

Furthermore, the interphase momentum exchange source term and the interphase energy exchange source term can be expressed as:

$$F_{gp} = -\iiint f_p \left\{ m_p \left[D_p \left(\boldsymbol{u}_g - \boldsymbol{u}_p \right) - \frac{\nabla p_g}{\rho_p} \right] + \boldsymbol{u}_p \frac{dm_p}{dt} \right\} dm_p d\boldsymbol{u}_p dT_p$$
(17)
$$S_{gp} = \iiint f_p \left\{ m_p \left[D_p \left(\boldsymbol{u}_g - \boldsymbol{u}_p \right)^2 - C_p \frac{dT_p}{dt} \right] - \frac{dm_p}{dt} \left[h_p + \frac{1}{2} \left(\boldsymbol{u}_g - \boldsymbol{u}_p \right)^2 \right] \right\} dm_p d\boldsymbol{u}_p dT_p$$
(18)

2.2. Chemical reaction model

After entering the gasifier, the moisture in biomass particles is first evaporated at high temperatures. The drying process is expressed as follows:

Moisture in biomass
$$\rightarrow$$
 H₂O (g) (R2)

According to the Arrhenius law, the volatilization rate of water is given by [37]:

$$r_{drying} = 5.13 \times 10^{10} \exp(-\frac{10585}{T_p}) m_{Moisture}$$
 (19)

where $m_{Moisture}$ is the mass of moisture in the biomass particle.

As the particle temperature increases, the biomass particles undergo pyrolysis, resulting in the production of fixed carbon, combustible gas, and tar. The main process of biomass particles pyrolysis can be expressed as:

Dry Biomass $\rightarrow \alpha_1 H_2 + \alpha_2 H_2 O + \alpha_3 CO + \alpha_4 CO_2 + \alpha_5 CH_4 + \alpha_6 Char + \alpha_7 Ash$ (R3)

where α (i = 1, 2, 3......7) represents the mass fraction of each pyrolysis product, and satisfies $\sum_i \alpha_i = 1$. According to the Arrhenius law, the reaction rate of the pyrolysis process is given by [7,38]:

$$\frac{dm_{volatiles}}{dt} = -5 \times 10^6 \exp(-\frac{1.44 \times 10^7}{T_p}) m_{volatile}$$
⁽²⁰⁾

where $m_{volatile}$ is the mass of the volatile.

There are also numerous complicated homogeneous and heterogeneous reactions within the gasifier and combustor. Achieving a comprehensive representation of all these reactions in the simulation process is a challenging task, thus necessitating some necessary simplifications. In the AER gasification process, the bed material CaO also catalyzes tar reforming which helps in further reducing the tar content in the product gas [39]. However, in the current work, the processes of tar generation and cracking are not taken into account. Additionally, trace elements such as N, S, and Cl are also disregarded in the calculation. Only the generation of CH_4 is considered for simplicity. It is worth mentioning that this simplification approach is commonly adopted by other researchers [7,40–42], enabling easier modeling and analysis. In

(a)



Fig. 9. The gas-solid flow pattern in the DFB reactor: (a) conventional gasification; (b) AER gasification.





addition, rapeseed methyl ester (RME) is added to the combustor as an additional fuel to provide heat during operation, and the main component is simplified by $C_{16}H_{34}$. The main homogeneous and heterogeneous reactions and reaction rates considered in this study are detailed in Table 1.

In the biomass AER gasification process, there are also carbonation reactions of CaO and calcination reactions of CaCO₃:

$$CaO + CO_2 \leftrightarrows CaCO_3, \Delta H_{298} = -178 \text{KJ/mol}$$
(R11)

The carbonation reaction (forward) is exothermic while the calcination reaction (reverse) is endothermic. Therefore, the shift of the reaction equilibrium is determined by the temperature and CO₂ partial pressure (P_{CO2}) in the reactor. The equilibrium partial pressure of CO₂ (P_{CO2} , e_q) as a function of temperature is expressed by Baker below [45,46], and the equilibrium diagram is shown in Fig. 2:

$$\log_{10} P_{\cos_2, eq} = 7.079 - \frac{8308}{T_g} \tag{21}$$

The carbon dioxide adsorption rate equation for CaO sorbent is referred by Sun et al. [47]:

$$R_0 = 56k_0(1 - X_0) \left(P_{CO_2} - P_{CO_2, eq} \right)^n \cdot A_0$$
(22)

where 56 represents the molar mass of CaO in g/mol. X_0 is the conversion rate of CaO and *n* is the reaction order. A_0 is the specific surface area



Fig. 10. Axial distributions of solid holdup in the reactors under conventional gasification and AER gasification conditions: (a) combustor; (b) gasifier.

of the CaO particle. k_0 is the reaction rate coefficient as:

$$k_0 = 1.67 \times 10^{-4} \exp\left(\frac{-E}{RT}\right), E = 29 \pm 4\text{KJ/mol}, n = 1, P_{CO_2} - P_{CO_2,eq}$$

$$\leq 10\text{KPa}$$
(23)

$$k_0 = 1.67 \times 10^{-3} \exp\left(\frac{-E}{RT}\right), E = 29 \pm 4\text{KJ/mol}, n = 0, P_{CO_2} - P_{CO_2,eq}$$

> 10KPa (24)

The reaction rate of the $CaCO_3$ calcination reaction in the combustor is expressed as [48]:

$$R_1 = k_1 (1 - X_1)^{2/3} \left(1 - \frac{P_{CO_2}}{P_{CO_2,eq}} \right)^{1.86}$$
(25)

where X_1 is the conversion rate of CaCO₃. k_1 is the reaction rate coefficient, given by:

$$k_1 = 5.61 \times 10^5 \exp\left(-\frac{150000}{RT}\right)$$
 (26)

3. Computational setting and model validation

3.1. Geometry and numerical setting

The numerically studied DFB gasification system is part of a 100 KWth DFB pilot plant at the Vienna University of Technology [49]. The schematic diagram and geometry structure of the DFB gasifier are schematically shown in Fig. 3(a, b) [50,51]. Specifically, the whole system is composed of a gasifier and a combustor which are connected by a solid separator, an upper loop-seal, and a bottom loop-seal.

The gasifier is a bubbling fluidized bed (BFB) composed of a square column and a cone. The side length of the square column section is 0.265 m, the equivalent diameter is 0.304 m, and the height is 2.35 m.

Steam is introduced as the fluidizing gas and gasification agent. After being introduced into the gasifier, the biomass particles first undergo a process of rapid drving and pyrolysis under high temperature. Then the pyrolysis gas reacts with char or steam in the gasifier. The final product gas overflows from the upper outlet of the gasifier. The remaining char particles and a part of the bed material are moved toward the combustor through the bottom loop-seal. The combustor is a circulating fluidized bed (CFB) comprised of a cylindrical standpipe with a height of 3.9 m and a diameter of 0.098 m. Air is utilized as the fluidizing gas, with primary air and secondary air being introduced from the gas nozzles located at the bottom and expansion part of the combustor, respectively. In order to provide combustion support and heat the bed material, RME is added as an additional fuel in the lower section of the combustor. Flue gas and bed material particles are effectively separated by a solid separator positioned at the top of the combustor. This device also ensures that the flue gas, along with the smaller-sized ash particles, can be efficiently discharged from the upper section of the solid separator. Meanwhile, the high-temperature bed material particles are intercepted and dropped through the baffle integrated within the separator. Then particles enter the gasifier through the upper loop-seal to provide heat for the gasification reaction. The steam-sealed loop-seal can prevent flue gas from entering the gasification reactor.

In the current work, the conventional gasification and AER gasification processes of biomass in DFB are numerically studied. The operation temperature of the gasifier and the combustor are given in Table 2. The biomass raw material is wood pellets with a density of 850 kg/m³. Table 3 details its ultimate analysis and proximate analysis [52]. Under the conventional gasification condition, the bed material is olivine, with a particle size distribution of $0.45 \sim 0.63$ mm and a density of 3300 kg/m³, and its compositions are given in Table 4 [53]. Under the AER gasification condition, the bed material used is calcined calcite, and its compositions are shown in Table 5 [51]. The PSD of calcite particles is $0.5 \sim 1.3$ mm and the density is 2800 kg/m³, which is consistent with the experimental measurement [51]. The PSD of the three kinds of



Fig. 11. Particle velocity distribution in the DFB under the conventional gasification condition (top) and AER gasification condition (bottom): (a, c) radial velocity; (b, d) vertical velocity.

particles all follow the normal distribution, which is consistent with the experimental measurement [54], as shown in Fig. 4.

The commercial software Barracuda Virtual Reactor is employed to perform the simulation. For the simulations, the settings of initial conditions and boundary conditions are determined according to actual operating parameters [49,55]. At the initial time, the total mass of bed material particles inside the DFB is 100 kg. The numbers of computational particles are 201,514 and 292,756 under conventional gasification and AER gasification conditions. Due to the greater particle density and smaller particle size of the conventional gasification bed material



Fig. 12. Particle species distribution in the DFB: (a) conventional gasification; (b) AER gasification.

compared to AER gasification, each parcel represents a real number of particles of 1757 and 790, respectively. And two loop-seals are filled with bed material and fluidized with steam. The inlets are set as the velocity inlet boundary with the fixed temperatures and the outlets are set as the pressure outlet boundary with the zero-gradient temperature. Meanwhile, the walls are set as the no-slip adiabatic boundary. The physical time of the simulation for each case is 100 s. The main operating parameters are summarized in Table 6.

In this work, four sets of gasification temperatures (600 °C, 650 °C, 700 °C, and 750 °C) and four sets of steam flow rates (0.00329 kg/s, 0.00439 kg/s, 0.00549 kg/s, and 0.00658 kg/s) are set to study their effects on the gasification performance under the AER gasification condition. In addition, four groups of absorbent content (25 %, 50 %, 75 %, and 100 %) are set up to study the relationship between absorbent content and gasification performance. Each simulation case was performed on a cluster outfitted with 16 CPU processors and a GeForce RTX 4060 GPU. The computational expense of the base case under the AER gasification condition is about 78 h in this work. The specific operating parameters are given in Table 7.

In addition, the computational domain is divided into grids with different resolutions: coarse grid (i.e., $N_c = 80712$), medium grid (i.e., $N_c = 139104$), and fine grid (i.e., $N_c = 240295$). The spatial distribution of time-average temperature can illustrate the heat transfer mechanisms in the reactor. Besides, the thermochemical behavior of biomass gasification in the DFB is the main concern of the present work. Thus, we choose a temperature distribution to convince that the thermal quantity is insensitive to the grid resolution, as shown in Fig. 5. In addition to the time-averaged pressure distribution, we have also added the axial distribution of time-averaged pressure under different grid resolutions in the reactors (see Fig. S1 of the Supporting Information). The results show that changing the grid resolution. Thus, the physical quantity is insensitive to the grid resolution. In Fig. 5, it can be observed that the

temperature distribution is significantly different under the coarse grid compared to the other two grids, whereas the temperature distribution under the medium and fine grids shows similarity. Based on the consideration of balancing calculation accuracy and amount, the medium grid is adopted in the following numerical simulations. As shown in Fig. 3(c), the computational domain is divided into structured grids by the Cartesian meshing method and the boundary grids are formed by cutting the structured grids using the actual geometry. At the same time, some regions are refined such as near-wall surfaces and component junctions. The average cell size is 0.02 mm \times 0.02 mm.

3.2. Model validation

Fig. 6 shows the variation in the mass fractions of gas products over time under conventional gasification and AER gasification conditions. It can be seen that the mass fractions of gas products increase rapidly in the initial start-up stage of the first 10 s, and fluctuate around fixed values after t = 50 s, indicating that the thermochemical behavior in the DFB reaches the steady state within the statistical time range. Therefore, the data after 50 s can be employed for time averaging. As shown in Fig. 7, the current model can well predict the gas composition both under conventional gasification and AER gasification conditions, and the error between the simulation results and experimental data [55] is within an acceptable range. The slight difference may be caused by the simplification of reactions and the use of empirical parameters of reaction kinetics. Therefore, we believe that the developed model can reasonably predict the chemical reaction behavior in the DFB system under the two gasification conditions. Fig. 8 shows the comparison of the simulation results and experimental data of the time-average temperature of the gasifier and combustor after reaching the steady state under the two gasification conditions. It can be seen that the simulated temperatures agree well with the experimental data, indicating that the established model can reasonably predict the heat transfer behavior in the reactors.



Fig. 13. Temperature distribution in the DFB under the conventional gasification condition (top) and AER gasification condition (bottom): (a, b) particle temperature; (c, d) gas phase temperature.

4. Result and discussion

4.1. Comparative analysis of two gasification processes

4.1.1. Gas-solid flow in the reactor

Fig. 9 illustrates the evolution of the gas-solid flow pattern under two gasification conditions in the DFB. In the initial state, a certain number of bed material particles are accumulated in the combustor, gasifier, and loop-seals. Air and steam are respectively introduced into the combustor and gasifier from the bottom of reactors. Due to the drag force, the static particles in the reactor experience an upward movement under the entrainment of gas. Conversely, the particles that have accumulated in the upper loop-seal slowly descend due to the influence of the airflow from the combustor and the return air, and some particles enter the gasifier. At the same time, biomass and additional fuel are introduced into the reactors from the feed port and mixed with the bed



Fig. 14. Axial distribution of gas phase temperature in the reactors under conventional gasification and AER gasification conditions: (a) combustor; (b) gasifier.

material particles, respectively. Under the conventional gasification condition, it can be observed that certain particles in the combustor reach the top of the riser at about 1 s. This part of the particles subsequently descends into the upper loop-seal due to the influence of the baffle. The ash with low density escapes through the exhaust pipe with the exhaust gas. At about 5 s, the particles that accumulated in the upper loop-seal have reached the maximum capacity. Then the bed material particles initiate a continuous descent into the gasifier. Particles in the bubbling bed are also conveyed into the combustor through the lower loop-seal. After about 20 s, the conventional gasification system realizes the full cycle and achieves dynamic stability. In contrast, the AER gasification system achieves stability in a relatively shorter time. Notably, particle accumulation reaches the maximum at approximately 2.5 s, while the gas-solid circulation attains dynamic stability at around 10 s. In the steady state stage, the combustor is in a rapid fluidization state, the gasifier is in a bubbling fluidization state, and the gas-solid flow pattern does not change greatly. However, compared with conventional gasification, under the AER gasification condition, fewer particles are transported from the combustor to the gasifier, and fewer bubbles are bubbled in the gasifier after stabilization.

Fig. 10 presents the axial distributions of the time-average solid holdup in the combustor and the gasifier under two gasification conditions. Under the conventional gasification condition, in the combustor, the solid holdup decreases first because the primary air enters from the bottom. Then, due to the bed material being circulated through the lower loop-seal from the gasifier at 0.2 m, the solid holdup in the riser increases. At a height of 0.4 m, the injection of secondary air from the lower part results in the solid content being characterized by an initial decrease followed by an increase. Then, the solid holdup gradually decreases with the increase in height, and the overall distribution is thicker at the bottom and thinner at the top. In the gasifier, the solid holdup decreases slightly at the steam and biomass feed and increases slightly at 2 m due to the bed material circulation in the combustor. Furthermore, a sharp drop of the solid holdup can be observed on the bed surface, approaching zero. As a result, a distinct transition from the dense phase region to the dilute phase region exists within the gasifier. Compared with the conventional gasification, the axial distribution trend of solid holdup under the AER gasification condition is similar. However, it can be seen from Fig. 10(b) that the bed height under the AER gasification condition is larger than that under the conventional gasification condition during stable operation. This is because the initial bed height in the AER gasifier is larger for the same initial mass, which is caused by the lower density of the bed material. Therefore, the effect of initial bed height is greater than the effect of bed expansion due to fluidization.

Fig. 11 presents the particle velocity in the DFB system under two gasification conditions. It is evident that the particle vertical velocity is one order of magnitude larger than the particle radial velocity under the two gasification conditions. Furthermore, the combustor exhibits significantly higher axial velocities for particles compared to the gasifier. In the gasifier, the particles with larger axial velocity are mainly concentrated on the surface of the bed or near the side wall where the biomass is fed. Compared to the conventional gasification, the AER gasification exhibits a smaller particle velocity, resulting in a gentler particle movement. Fig. 12 shows the solid species distribution in the DFB system under two gasification conditions. On the bed surface of the gasifier, particle segregation can be observed due to the difference in mass or density, and biomass particles are mainly distributed on the bed surface. However, compared to the conventional gasification condition, under the AER gasification condition, particle movement is intensified. This results in a more dispersed distribution of biomass particles on the bed surface, and the bed material becomes more thoroughly mixed under the conventional gasification condition.

4.1.2. Temperature distribution in the reactor

Fig. 13 shows the spatial distribution of instantaneous particle phase temperature and time-averaged gas phase temperature in the DFB under conventional gasification and AER gasification conditions. Due to the different chemical reactions in two reactors, the exothermic reaction between the fuel and carbon in the combustor elevates the temperature, while the endothermic gasification reaction in the gasifier decreases the



Fig. 15. Comparison of the contours of gas species concentrations in the DFB under the conventional gasification condition (left) and AER gasification condition (right): (a) CO; (b) CO₂; (c) CH₄; (d) H₂; (e) H₂O; (f) O₂.



(b)

Fig. 16. Axial distribution of gas species in the reactors under the conventional gasification (left) and AER gasification (right) conditions: (a) combustor; (b) gasifier.

temperature, resulting in a temperature disparity between the two reactors. Under the two gasification conditions, the low-temperature area at the bottom of the combustor is formed due to the entry of lowtemperature primary air and circulating particles. The secondary air in the expansion section of the riser accelerates the combustion reaction of the additional fuel and char, consequently causing a rapid increase in temperature. Subsequently, the high-temperature particles enter the gasifier through the upper loop-seal, providing the requisite energy for the gasification process. In the gasifier, the local low-temperature areas are mainly concentrated at the steam inlet and biomass feed. Under the conventional gasification condition, bed material particles serve as heat carriers between reactors. In contrast, under the AER gasification condition, bed material particles also function as $\rm CO_2$ carriers, facilitating the transfer of $\rm CO_2$ from the gasifier to the combustor. Furthermore, compared to the conventional gasification condition, under the AER gasification condition, the temperature difference between reactors is



Fig. 17. Low heating value and combustible gas concentration of gas products under the conventional gasification and AER gasification conditions.

larger, and the presence of heat flow particles in the gasifier is more obvious.

Fig. 14 shows the axial time-average temperature distribution of the gas phase in the reactors under the two gasification conditions. In the combustor, the temperature initially increases with the bed height increasing due to the combustion reaction between the primary air and the remaining char. At the bottom loop-seal, due to the low-temperature gas and particles moving into the gasifier, the temperature decreases sharply. Subsequently, as additional fuel enters, the combustion reaction occurs that releases more heat, causing the temperature to rise continuously along the axial height. With the entry of secondary air and the complete mixing of gas and solid, the temperature gradually stabilizes. In the gasifier, the temperature of the surrounding bed layer drops slightly due to the entry of the low-temperature gasification agent. Above the bed surface, there is a localized high temperature influenced by the high-temperature particles transported from the combustor. Compared with conventional gasification, the temperature in the reactors under the AER gasification condition is lower. Additionally, due to the larger temperature difference in the reactors, heat transfer is more pronounced, and temperature fluctuations are more severe under the AER gasification condition. Simultaneously, in the gasifier, the



Fig. 18. Effect of gasification temperature on gas species (a) and the LHV and CGC (b) under the AER gasification condition.



Fig. 19. Effect of steam flow rate on gas species (a) and LHV and CGC (b) under the AER gasification condition.



Fig. 20. Effect of absorbent content on gas species (a) and LHV and CGC (b) under the AER gasification condition.

absorbent of CO_2 is an exothermic reaction, causing the temperature to increase at about 1 m.

4.1.3. Gasification products

Fig. 15 shows the contours of gas species concentrations in the DFB system under two gasification conditions. Since there are different chemical reaction processes taking place in the combustor and the gasifier, the gas composition in these reactors is different. Under the conventional gasification condition, the combustion reaction of additional fuel and remaining char is mainly carried out in the combustor. As the height increases, the O_2 concentration gradually decreases, while the CO_2 concentration gradually increases. Furthermore, it can be observed that the upper and lower loop-seals between the two reactors are filled with steam, which effectively prevents the flue gas in the combustor from infiltrating the gasifier.

The pyrolysis of biomass particles and the gasification reaction of char particles are mainly carried out in the gasifier. As the inlet of the fuel particles, the biomass particles are rapidly heated by the high-temperature gas phase to release volatile gases such as CO, CO₂, H₂, and CH₄. Therefore, a high concentration of gases, such as CO, can be observed near the fuel inlet. Simultaneously, the introduction of steam enhances the water–gas shift reaction and steam gasification reaction, thereby increasing the concentration of H₂. Compared with conventional gasification, the carbonation reaction of CO₂ and CaO takes place under the AER gasification condition. This significantly decreases the CO₂ concentration of CH₄ and CO within the gasifier diminishes, leading to the production of gas with a high H₂ content.

To obtain a deeper understanding of the gas composition profile, the axial concentration distributions of four main gas components, CO, CO₂, H₂, and H₂O are presented in Fig. 16. As the height increases, it can be observed that H₂O first enters the lower loop-seal, and its concentration gradually decreases with the entry of the secondary air in the combustors. Subsequently, additional fuel enters and reacts with O₂, leading to a gradual increase in CO₂ content, while the H₂ and CO content remain essentially zero. In the gasifier, the concentrations of CO, H₂, and CO₂ gradually increase due to the pyrolysis of biomass particles. Additionally, there is a rapid drop in the concentration of H₂O on the surface of the bed, accompanied by a rapid rise in the concentration predominantly occurs on the bed surface. At about 2.1 m, the H₂O in the upper loop-seal enters the gasifier with the circulating bed material, causing fluctuations in gas concentration, which eventually stabilizes with the reaction

equilibrium. Compared with conventional gasification, the contents of CO and CO_2 in the gasifier are smaller under the AER gasification condition. This indicates that the adsorption process has a greater impact on the related parallel reforming/gasification reactions.

It can be seen from Fig. 7 that compared with conventional gasification, AER gasification reduces the CO_2 mole fraction by 76.67 % and increases the H₂ mole fraction by 54.58 %. This work also compares the lower heating value (LHV) and combustible gas (H₂, CO, CH₄) concentration (CGC) of the gas products of the DFB system under two gasification conditions, as shown in Fig. 17. The lower heating value and combustible gas concentration are respectively defined as [44,56]:

LHV
$$(MJ/Nm^3) = (25.7 \times H_2\% + 30.3 \times CO\% + 85.4 \times CH_4\%) \times (4.2 / 1000)$$
 (27)

CGC (%) = The volume summation of H_2 , CO and CH_4 in syngas / Total volume of syngas (without H_2O) × 100% (28)

Under the conventional gasification and AER gasification conditions, the LHV of gas products are 11.20 MJ/Nm^3 and 13.76 MJ/Nm^3 , and the CGC is 78.89 % and 95.07 %, respectively. Thus, AER gasification can significantly improve the quality of gas products.

4.2. Effect of operating parameters on the gasification process

4.2.1. Effect of gasification temperature on the gasification process

In this section, the effect of some key operating parameters on the AER biomass gasification process is detailed discussed. Fig. 18(a) shows the gas composition under different operating temperatures and it can be seen that the mole fraction of H2 increases gradually from 72.19 % to 77.50 % with the gasification temperature increasing from 600 $^\circ \mathrm{C}$ to 750 °C. At the same time, the mole fraction of other gas species decreases, in which the CO mole fraction decreases most obviously, from 8.75 % to 5.89 %. This is attributed to the enhanced carbonation reactions of CO2 and CaO, as well as the methane steam reforming reaction. Therefore, the decrease of CO₂ and CH₄ mole fractions affects the water-gas shift reaction, shifting it towards the production of H₂. Fig. 18 (b) illustrates the changes of CGC and LHV under different gasification temperatures. With the increase of gasification temperature, LHV first increases and reaches the maximum at 650 °C. Then, the LHV decreases gradually due to the decrease of the mole fractions of CH₄ and CO with high calorific values in the gas products. On the other hand, CGC increases gradually with the increase of gasification temperature.

4.2.2. Effect of steam flow rate on the gasification process

Fig. 19(a) illustrates the influence of the steam flow rate in the gasifier on the gas species under the AER gasification condition. It can be observed that with the increase of the steam flow rate, the mole fraction of CO₂ first decreases and then increases, while the changes of CO and H₂ have no certain rules. The increase in steam flow affects both the gas-solid dynamics and chemical reaction in the gasifier simultaneously. When the steam flow rate increases from 0.00329 kg/s to 0.00439 kg/s, the bubbling phenomenon in the reactor becomes more apparent, resulting in better solid mixing and increased gas-solid contact area. When the steam flow rate increases from 0.00329 kg/s to 0.00439 kg/s, the bubbling phenomenon in the furnace is more obvious, the solid mixing is more sufficient, and the gas-solid contact area increases. Therefore, more CO₂ can be absorbed, and the increase of steam promotes the water-gas shift reaction, resulting in the decrease of the CO mole fraction and the increase of the H₂ mole fraction. As the steam flow is further increased, the residence time of the gases in the furnace decreases. The water-gas shift reaction has not yet reached equilibrium, resulting in an increase in the CO mole fraction and a decrease in the H₂ mole fraction. However, when the steam flow rate is 0.00659 kg/s, the effect of steam increase on the water-gas shift reaction rate becomes the dominant factor, accelerating the conversion of CO to H₂. It can be seen from Fig. 19(b) that when the steam flow rate reaches 0.00439 kg/s, the increase of steam has no obvious effect on the improvement of LHV and CGC. Therefore, increasing the steam flow rate has no significant improvement on the gasification performance.

4.2.3. Effect of absorbent content on the gasification process

To better understand the influence of the absorbent content on the biomass gasification process, four groups of bed materials with different absorbent contents are set up, which are 25 % calcined calcite and 75 %olivine, 50 % calcined calcite and 50 % olivine, 75 % calcined calcite and 25 % olivine, and 100 % calcined calcite, respectively. It can be seen from Fig. 20(a) that with the increase of the absorbent content, the mole fractions of CH₄ and H₂ increase, and the mole fraction of CO decreases. Comparable, the mole fraction of CO₂ decreases greatly at first and then increases slightly. As the content of calcined calcite increases from 25 % to 75 %, the contact area between the gas and the absorbent increases. This leads to a reduction in CO₂ mole fraction and enhances the water-gas shift reaction, ultimately resulting in a decrease in CO mole fraction and an increase in H2 mole fraction. When the absorbent content reaches 100 %, the dominant factor is the decrease in solids circulation rate. This results in a reduction of heat transportation to the gasifier through the bed material particles, which weakens the methane steam reforming and carbonation reaction, and increases the water-gas shift reaction. Consequently, the mole fractions of CO₂, CH₄, and H₂ increase, while the mole fraction of CO decreases. It can be seen from Fig. 20(b) that with the increase of absorbent content, both LHV and CGC increase first and then decrease. The LHV reaches the maximum when the absorbent content is 75 % due to the higher calorific value of CO compared to H₂. At the same time, CGC is also the maximum value because of the lowest CO2 content. Therefore, the cost of bed material can be reduced by properly adjusting the content of absorbent.

5. Conclusion

In this work, a 100 KWth DFB reactor is numerically studied under the frame of the MP-PIC method and both the conventional gasification and AER gasification conditions are investigated. Furthermore, the thermochemical behavior such as heat transfer and complex reaction kinetics of gasification, carbonation, and calcination are all considered. Firstly, the simulation results are compared with the experimental data to verify the accuracy of the developed model. Then the comparative analysis of the flow behavior and reaction characteristics in the DFB system under two gasification conditions is detailed discussed. Finally, the effects of several key operating parameters, such as gasification temperature, steam flow rate, and absorbent content on the gasification performance are investigated under the AER gasification condition. The main conclusions of this study are as follows:

- (1) The reaction kinetics considering carbonation and calcination reactions are confirmed to be reliable in describing the conventional gasification and AER gasification processes. Compared with conventional gasification, AER gasification reduces the CO_2 mole fraction by 76.67 %, increases the H₂ mole fraction by 54.58 %, and greatly increases the hydrogen content in the product gas. Under conventional gasification and AER gasification and AER gasification conditions, the LHV of gas products are 11.2 MJ/Nm³ and 13.76 MJ/Nm³, and the CGC is 78.89 % and 95.07 %, respectively. It shows that the AER gasification can greatly improve the quality of export gas.
- (2) Compared with the conventional gasification, the particle movement in the reactor is gentler, the bed expansion height is lower, and the temperature difference between the two reactors is larger under the AER gasification condition. Furthermore, the temperature of the reactor is also lower, which is beneficial to the carbonation reaction. In the gasifier, due to the difference in particle size and density, segregation occurs, and most of the biomass particles are distributed on the bed surface. In addition, the gasification process mainly occurs in the gasifier near the inlet side. Compared with the conventional gasification, the content of CO and CO₂ in the gasifier is lower under the AER gasification condition. This shows that the adsorption process has a greater impact on the related parallel reforming/gasification reactions.
- (3) Higher temperature can improve the performance of AER gasification, while the effect of steam flow rate on gasification performance depends on many factors. With the increase in gasification temperature, the carbonation and methane steam reforming reactions are enhanced. As a result, the CO mole fraction decreases, and the H₂ mole fraction increases. The increase in steam flow affects the gas–solid flow and various chemical reactions in the gasifier at the same time. Therefore, the gasification performance can only be improved when the promotion effect of steam on the water–gas shift reaction is dominant. The higher steam flow rate cannot significantly improve the gasification performance under the AER gasification condition.
- (4) Properly adjusting the content of absorbent can reduce the cost of bed materials. With the increase of the absorbent content in the bed material, the mole fractions of CH₄ and H₂ increase, the mole fraction of CO decreases, and the mole fraction of CO₂ decreases first and then increases. The LHV and CGC of the gas products are the maximum when the absorbent content is 75 %.

The numerical results in this study can help better understand the hydrodynamics and thermochemical characteristics of the AER biomass gasification in the DFB.

CRediT authorship contribution statement

Mengyao Guo: Validation, Visualization, Writing – original draft. Jiahui Yu: Writing – review & editing. Shuai Wang: Writing – review & editing. Kun Luo: Writing – review & editing. Jianren Fan: Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data that has been used is confidential.

Acknowledgments

We are grateful for the support from the National Natural Science Foundation of China (grant No. 51925603) and the Fundamental Research Funds for the Central Universities (2022ZFJH004).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2024.148723.

References

- Y. Cheng, Z. Thow, C. Wang, Biomass gasification with co2 in a fluidized bed, Powder Technol. 296 (2016) 87–101.
- [2] F. Saleem, J. Harris, K. Zhang, Non-thermal plasma as a promising route for the removal of tar from the product gas of biomass gasification-a critical review, Chem. Eng. J. 1 (2020) 382.
- [3] S.Y. Foong, R.K. Liew, Y. Yang, Y.W. Cheng, P.N.Y. Yek, W.A.W. Mahari, et al., Valorization of biomass waste to engineered activated biochar by microwave pyrolysis: progress, challenges, and future directions, Chem. Eng. J. 389 (2020) 124401.
- [4] D. Kong, S. Wang, K. Luo, Q. Xu, J. Fan, Insights of biomass gasification combined with co2 absorption enhanced reforming in an 8 mwth dual fluidized bed, Chem. Eng. J. 466 (2023) 142981.
- [5] M.J. Prins, K.J. Ptasinski, F.J. Janssen, From coal to biomass gasification: comparison of thermodynamic efficiency, Energy (Oxf) 32 (7) (2007) 1248–1259.
- [6] A. Molino, S. Chianese, D. Musmarra, Biomass gasification technology: the state of the art overview, J. Energy Chem. 25 (1) (2016) 10–25.
- [7] X. Ku, T. Li, T. Løvås, Cfd-dem simulation of biomass gasification with steam in a fluidized bed reactor, Chem. Eng. Sci. 122 (2015) 270–283.
- [8] Beohar H, Gupta B, Sethi VK, Pandey M. Parametric study of fixed bed biomass gasifier: a review 2012.
- [9] J. Zhou, Q. Chen, H. Zhao, X. Cao, Q. Mei, Z. Luo, et al., Biomass-oxygen gasification in a high-temperature entrained-flow gasifier, Biotechnol. Adv. 27 (5) (2009) 606–611.
- [10] B. Feng, H. An, E. Tan, Screening of co2 adsorbing materials for zero emission power generation systems, Energy Fuels 21 (2) (2007) 426–434.
- [11] A.M. Parvez, S. Hafner, M. Hornberger, M. Schmid, G. Scheffknecht, Sorption enhanced gasification (seg) of biomass for tailored syngas production with in-situ co2 capture: current status, process scale-up experiences and outlook, Renew. Sustain. Energy Rev. 141 (2021) 110756.
- [12] F.N. Ridha, Y. Wu, V. Manovic, A. Macchi, E.J. Anthony, Enhanced co2 capture by biomass-templated ca (oh) 2-based pellets, Chem. Eng. J. 274 (2015) 69–75.
- [13] N. Hanchate, S. Ramani, C.S. Mathpati, V.H. Dalvi, Biomass gasification using dual fluidized bed gasification systems: a review, J. Clean. Prod. 280 (2021) 123148.
- [14] J. Karl, T. Pröll, Steam gasification of biomass in dual fluidized bed gasifiers: a review, Renew. Sustain. Energy Rev. 98 (2018) 64–78.
- [15] Fuchs J, Schmid JC, Mueller S, Hofbauer H. Dual fluidized bed gasification of biomass with selective carbon dioxide removal and limestone as bed material: a review. Renew. Sustain. Energy Rev. 2019;107(JUN.):212-31.
- [16] S. Müller, J. Fuchs, J.C. Schmid, F. Benedikt, H. Hofbauer, Experimental development of sorption enhanced reforming by the use of an advanced gasification test plant, Int. J. Hydrogen Energy 42 (50) (2017) 29694–29707.
- S. Koppatz, C. Pfeifer, R. Rauch, H. Hofbauer, T. Marquard-Moellenstedt,
 M. Specht, H2 rich product gas by steam gasification of biomass with in situ co2 absorption in a dual fluidized bed system of 8 mw fuel input, Fuel Process. Technol. 90 (7–8) (2009) 914–921.
- [18] N. Poboss, K. Swiecki, A. Charitos, C. Hawthorne, M. Zieba, G. Scheffknecht, Experimental investigation of the absorption enhanced reforming of biomass in a 20 kwth dual fluidized bed system, Int. J. Thermodyn. 15 (1) (2012) 53–59.
- [19] F. Alobaid, N. Almohammed, M.M. Farid, J. May, P. Rößger, A. Richter, et al., Progress in cfd simulations of fluidized beds for chemical and energy process engineering, Prog. Energy Combust. Sci. 91 (2022) 100930.
- [20] H. Liu, R. Cattolica, J. Seiser, et al., Operating parameter effects on the solids circulation rate in the cfd simulation of a dual fluidized-bed gasification system, Chem. Eng. Ence (2017).
- [21] S. Kraft, F. Kirnbauer, H. Hofbauer, Cpfd simulations of an industrial-sized dual fluidized bed steam gasification system of biomass with 8mw fuel input, Appl. Energy 190 (2017) 408–420.
- [22] J. Wang, Continuum theory for dense gas-solid flow: a state-of-the-art review, Chem. Eng. Sci. 215 (2020) 115428.
- [23] R.I. Singh, A. Brink, M. Hupa, Cfd modeling to study fluidized bed combustion and gasification, Appl. Therm. Eng. 52 (2) (2013) 585–614.
- [24] S. Wang, K. Luo, C. Hu, L. Sun, J. Fan, Impact of operating parameters on biomass gasification in a fluidized bed reactor: an eulerian-lagrangian approach, Powder Technol. 333 (2018) 304–316.

- [25] C. Li, Q. Eri, Comparison between two eulerian-lagrangian methods: cfd-dem and mppic on the biomass gasification in a fluidized bed, Biomass Convers. Biorefin. (2021) 1–18.
- [26] H. Liu, R.J. Cattolica, R. Seiser, C. Liao, Three-dimensional full-loop simulation of a dual fluidized-bed biomass gasifier, Appl. Energy 160 (2015) 489–501.
- [27] H. Sun, G. Bao, S. Yang, J. Hu, H. Wang, Numerical study of the biomass gasification process in an industrial-scale dual fluidized bed gasifier with 8mwth input, Renew. Energy 211 (2023) 681–696.
- [28] J. Xie, W. Zhong, Y. Shao, G. Zhou, Simulation of co-gasification of coal and wood in a dual fluidized bed system, Adv. Powder Technol. 32 (1) (2021) 52–71.
- [29] D.M. Snider, S.M. Clark, P.J. O'Rourke, Eulerian–Jagrangian method for threedimensional thermal reacting flow with application to coal gasifiers, Chem. Eng. Sci. 66 (6) (2011) 1285–1295.
- [30] P.J. O'Rourke, D.M. Snider, Inclusion of collisional return-to-isotropy in the mp-pic method, Chem. Eng. Sci. 80 (2012) 39–54.
- [31] S.E. Harris, D.G. Crighton, Solitons, solitary waves, and voidage disturbances in gas-fluidized beds, J. Fluid Mech. 266 (1994) 243–276.
- [32] D. Gidaspow, Multiphase Flow and Fluidization: Continuum and Kinetic Theory Descriptions, Academic press, 1994.
- [33] S. Yin, W. Zhong, B. Jin, J. Fan, Modeling on the hydrodynamics of pressurized high-flux circulating fluidized beds (phfcfbs) by eulerian–lagrangian approach, Powder Technol. 259 (2014) 52–64.
- [34] X. Wang, B. Jin, W. Zhong, Y. Zhang, M. Song, Three-dimensional simulation of a coal gas fueled chemical looping combustion process, Int J Greenh Gas Control 5 (6) (2011) 1498–1506.
- [35] S. Kongkitisupchai, D. Gidaspow, Carbon dioxide capture using solid sorbents in a fluidized bed with reduced pressure regeneration in a downer, AIChE J 59 (12) (2013) 4519–4537.
- [36] W.E. Ranz, W.R. Marshall, Evaporation from droplets: part i and ii, Chem. Eng. Prog. 48 (4) (1952) 141–173.
- [37] J. Xu, L. Qiao, Mathematical modeling of coal gasification processes in a wellstirred reactor: effects of devolatilization and moisture content, Energy Fuels 26 (9) (2012) 5759–5768.
- [38] N. Prakash, T. Karunanithi, Kinetic modeling in biomass pyrolysis-a review, J. Appl. Sci. Res. 4 (12) (2008) 1627–1636.
- [39] J.C. Schmid, J. Fuchs, F. Benedikt, A.M. Mauerhofer, S. Müller, H. Hofbauer, et al., Sorption enhanced reforming with the novel dual fluidized bed test plant at tu wien, European Biomass Conference and Exhibition (EUBCE). Stockholm. (2017) 421–428.
- [40] S. Du, J. Wang, Y. Yu, Q. Zhou, Coarse-grained cfd-dem simulation of coal and biomass co-gasification process in a fluidized bed reactor: effects of particle size distribution and operating pressure, Renew. Energy 202 (2023) 483–498.
- [41] A. Ergüdenler, A.E. Ghaly, F. Hamdullahpur, A.M. Al-Tawel, Mathematical modeling of a fluidized bed straw gasifier: part i—model development, Energy Source. 19 (10) (1997) 1065–1084.
- [42] S. Gerber, F. Behrendt, M. Oevermann, An eulerian modeling approach of wood gasification in a bubbling fluidized bed reactor using char as bed material, Fuel (Lond) 89 (10) (2010) 2903–2917.
- [43] S. Sazhin, Modelling of heating, evaporation and ignition of fuel droplets: combined analytical, asymptotic and numerical analysis, J. Phys.: Conf. Series. IOP Publishing. (2005) 174.
- [44] D. Kong, S. Wang, K. Luo, C. Hu, D. Li, J. Fan, Three-dimensional simulation of biomass gasification in a full-loop pilot-scale dual fluidized bed with complex geometric structure, Renew. Energy 157 (2020) 466–481.
- [45] E.H. Baker, 87. The calcium oxide–carbon dioxide system in the pressure range 1–300 atmospheres, J. Chem. Soc. (resumed) (1962) 464–470.
- [46] K. Johnsen, J.R. Grace, S.S. Elnashaie, L. Kolbeinsen, D. Eriksen, Modeling of sorption-enhanced steam reforming in a dual fluidized bubbling bed reactor, Ind. Eng. Chem. Res. 45 (12) (2006) 4133–4144.
- [47] P. Sun, J.R. Grace, C.J. Lim, E.J. Anthony, Determination of intrinsic rate constants of the cao-co2 reaction, Chem. Eng. Sci. 63 (1) (2008) 47–56.
- [48] Z. Li, N. Cai, Modeling of multiple cycles for sorption-enhanced steam methane reforming and sorbent regeneration in fixed bed reactor, Energy Fuels 21 (5) (2007) 2909–2918.
- [49] C. Pfeifer, B. Puchner, H. Hofbauer, Comparison of dual fluidized bed steam gasification of biomass with and without selective transport of co2, Chem. Eng. Sci. 64 (23) (2009) 5073–5083.
- [50] S. Koppatz, C. Pfeifer, H. Hofbauer, Comparison of the performance behaviour of silica sand and olivine in a dual fluidised bed reactor system for steam gasification of biomass at pilot plant scale, Chem. Eng. J. 175 (2011) 468–483.
- [51] G. Soukup, C. Pfeifer, A. Kreuzeder, H. Hofbauer, In situ co2 capture in a dual fluidized bed biomass steam gasifier-bed material and fuel variation, Chem. Eng. Technol.: Industrial Chemistry-Plant Equipment-Process Engineering-Biotechnology 32 (3) (2009) 348–354.
- [52] S. Kern, C. Pfeifer, H. Hofbauer, Gasification of wood in a dual fluidized bed gasifier: influence of fuel feeding on process performance, Chem. Eng. Sci. 90 (2013) 284–298.
- [53] C. Pfeifer, R. Rauch, H. Hofbauer, In-bed catalytic tar reduction in a dual fluidized bed biomass steam gasifier, Ind. Eng. Chem. Res. 43 (7) (2004) 1634–1640.
- [54] R.K. Thapa, B.M. Halvorsen, C. Pfeifer, Modeling of reaction kinetics in bubbling fluidized bed biomass gasification reactor, Int. J. Energy Environ. (print) (2013) 5.
- [55] Höftberger E, Höftberger E. In-situ co2-adsorption in a dual fluidised bed biomass steam gasifier to produce a hydrogen rich gas: na; 2005.
- [56] C. Loha, H. Chattopadhyay, P.K. Chatterjee, Three dimensional kinetic modeling of fluidized bed biomass gasification, Chem. Eng. Sci. 109 (2014) 53–64.