



The influence of CO₂ gas concentration on the char temperature and conversion during oxy-fuel combustion in a fluidized bed

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HIGHLIGHTS

- Pyrometry with digital camera allows accurate measurement of char temperature in FB.
- Carbon gasification is significant even at a relatively low char temperature (850 °C).
- The char time out of the emulsion explains the different rate in O₂/CO₂ and O₂/N₂.
- The model takes into account explicitly the char particle's movement between phases.
- The model predicts and explains our own measurements and other from literature.

ARTICLE INFO

Keywords:

Fluidized bed
Oxy-combustion
Char temperature
Pyrometry
Coal
Biomass

ABSTRACT

In spite of the extensive theoretical and experimental work carried out on coal/char oxy-combustion in a fluidized bed (FB), the effect of changing the atmosphere from O₂/N₂ to O₂/CO₂ for a high O₂ concentrations is not entirely understood. In this work, experiments with single char particles are conducted in a bi-dimensional FB at 800 and 850 °C, varying the O₂ concentration from 11 to 50%_{v/v} in N₂ or CO₂. The FB reactor has a quartz window for visual observation, allowing the measurement of temperature and tracking the char conversion process by pyrometry with a digital camera. The method is shown to overcome the inherent limitations of other methods used in FB, such as thermocouples or pyrometry with an optical probe. Results indicate that the transfer of O₂ from the bulk gas of the bed to the surface of a char particle controls the overall rate of char conversion in O₂/N₂ and in O₂/CO₂. In the latter gas mixture, the carbon consumption by gasification is significant even at a relatively low char temperature (850 °C). This additional carbon consumption makes the apparent char consumption rate in both atmospheres roughly equal (at the same O₂ concentration) for char temperatures below 925 °C, and higher in O₂/CO₂ than in O₂/N₂ for char temperatures above 925 °C. Moreover, during the time in which the char stays in the emulsion phase, its temperature is roughly the same in both atmospheres, but when the char is in the bubble or splash zone its temperature is much higher than that in the emulsion phase. As a result, the difference in char conversion rate, observed in both atmospheres, is mainly controlled by the time in which the char particle is out of the emulsion phase. These results underline the importance of paying attention to the movement of a char particle through the different phases of the bed in order to improve the understanding of the oxy-fuel behavior in FB.

1. Introduction

More than 25% of the global electric energy demand is currently covered by coal combustion and an increase of coal utilization is expected, since it is the main energy source in some developing countries [1]. As a result, the need of reduction of CO₂ emission will need both increasing efficiency of power plants and the development of methods of CO₂ capture and storage [2]. Oxy-combustion is one of the proposed

technologies for this purpose with reasonable net efficiency and avoided CO₂ emissions [3,4]. In this process, coal conversion is carried out in an O₂ atmosphere diluted with CO₂ from recirculation of the flue gases [4] in order to control the conversion temperature and to reach a CO₂ concentration at the exit of the boiler ready for capture (> 90%) [5]. Furthermore, the NO_x and SO₂ emissions are reduced [6].

The first generation of oxy-boilers involves a retrofit of power plants where flue-gas recirculation maintains similar conditions inside the

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Nomenclature

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| $A_{\text{co/co2}}$ | pre-exponential factor for CO/CO ₂ ration, – |
| A_{cb} | pre-exponential factor for char combustion, m/Ks |
| A_{gf} | pre-exponential factor for char gasification, m/Ks |
| Ar | Archimedes number based on average inert particle diameter, – |
| c_p | specific heat of char particle, J/kg K |
| $c_{p,i}$ | specific heat of inert particles of the bed, J/kg K |
| $c_{p,g}$ | specific heat of fluidization gas, J/kg K |
| C | total molar concentration, mol/m ³ |
| C_d | dimensionless drag force of a isolated sphere, – |
| d_c | diameter of a char particle, m |
| d_i | average diameter of bed particles, m |
| d_{ij} | binary diffusivity of compound i through j, m ² /s |
| D_{ij} | effective mass diffusivity of compound i through j in the emulsion phase: $(\epsilon_{mf}/\tau)d_{ij}$, m ² /s |
| $E_{\text{co/co2}}$ | activation energy for CO/CO ₂ ratio, J/mol |
| E_{cb} | activation energy for char combustion, J/mol |
| E_{gf} | activation energy for char combustion, J/mol |
| h_{bed} | expanded bed height, m |
| h_p | penetration depth of a char particle in the bed, m |
| k_{cb} | external combustion kinetics, m/s |
| k_{gf} | external gasification kinetics, m/s |
| $k_{d,ePh}$ | effective thermal conductivity of emulsion phase, W/m K |
| $k_{d,i}$ | thermal conductivity of bed particles, W/m K |
| $k_{d,g}$ | thermal conductivity of the fluidization gas, W/m K |
| N_i | i-th molar flux around a char particle (>0 from the fluidized phases to the char surface), mol/m ² s |
| $N_{i,c}$ | i molar flux at the external surface of a char particle (>0 from the fluidized phases to the char surface), mol/m ² s |
| Nu | Nusselt number based on char particle diameter, – |
| Pr | Prandtl number: $c_{p,g} \mu_g / k_{d,g}$, – |
| $q_{\text{cv,g}}$ | heat exchange between the char particles and the fluidization gas by convection in Fig. 1 |
| $q_{\text{cv,b}}$ | heat exchange between the char particles and the inert particles of the bed by convection in Fig. 1 |
| $q_{\text{rd,b}}$ | heat exchange between the char particles and the inert particles of the bed by radiation in Fig. 1 |
| $q_{\text{cv,w}}$ | heat exchange between char particles and the wall of the FB reactor by radiation in Fig. 1 |
| r_c | radius of a char particle, m |
| $r_{\text{O2,cb}}$ | O ₂ molar flux consumed by oxidation at the external surface of a char particle, mol/m ² s |
| $r_{\text{O2,gf}}$ | CO ₂ molar flux consumed by gasification at the external surface of a char particle, mol/m ² s |
| R_g | gas constant: 8.315 J/mol K |
| Re_{ePh} | Reynolds number in the emulsion phase based on char particle size and u_{mf} , – |
| Re_{bPh} | Reynolds number in the bubble phase based on char particle size and u_{tf} , – |
| Re_{sPh} | Reynolds number in the splash zone based on char particle size and u_f , – |
| Sc | Schmidt number, $\mu_g / d_{ij} \rho_g$, – |
| Sh_{EMCD} | Sherwood number when there is equimolar counter-diffusion and the gas-film around the particle is not static: there is a convective flow |

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| Sh_o | Sherwood number when there is equimolar counter-diffusion and the gas-film around the particle is static |
| SUPM | Shrinking Unreactive Particle Model |
| t^* | characteristic time in which the char particle changes the temperature |
| T | time of a cycle of the movement of a char particle through the bed, s |
| $T_{j,T}$ | time of a char particle in the emulsion phase ($j = \text{ePh}$), bubble phase ($j = \text{bPh}$), or splash zone (sPh) during a cycle of its movement, s |
| T_c | combustion temperature of a char particle, K in equations and °C in figures |
| T_{bed} | bed temperature, K |
| u_b | average bubble velocity, m/s |
| u_d | average descent velocity of a char particle through the bed, m/s |
| u_f | fluidization velocity, m/s |
| u_{mf} | minimum fluidization velocity, m/s |
| u_r | average rise velocity of a char particle through the bed, m/s |
| u_{tf} | bubble through-flow velocity, m/s |
| $x_{c,fx}$ | content of carbon in the char particles in Eq. (29) |
| $x_{i,c}$ | molar concentration at the surface of a char particle, – |
| $x_{i,\text{bed}}$ | molar concentration in the fluidized phases, – |
| z | dimensionless parameter in AI:4, – |

Greek letters

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| Γ_c | apparent carbon consumption in Eq. (29), g/m ² s |
| ΔH_{cb} | heat of combustion (exothermic), J/mol |
| ΔH_{gf} | heat of gasification (endothermic), J/mol |
| δ | thickness of the gas-film around a char particle that absorbs the mass transfer, m |
| ϵ_{mf} | bed porosity at minimum fluidization velocity, – |
| θ | correction factor when there is no equimolar counter-diffusion, Eq. (7) |
| μ_g | viscosity of the fluidization gas, kg/ms |
| ξ | CO/CO ₂ ratio at the char surface by oxidation, – |
| ρ_b | density of the bed, kg/m ³ |
| ρ_c | density of a char particle, kg/m ³ |
| ρ_g | density of the fluidization gas, kg/m ³ |
| ρ_i | density of an inert particle of the bed, kg/m ³ |
| ρ_{mc} | molar density of a char particle, mol/m ³ |
| σ | Stefan-Boltzmann constant, $5.67 \cdot 10^{-8} \text{ W/m}^2 \text{ K}^4$ |
| ω_c | surface emissivity of a char particle |
| τ | tortuosity in Eq. (6) |
| Ψ_j | dimensionless parameter in AI:3, $j = \text{ePh}$ and $j = \text{bPh}$ |

Abbreviations

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| bPh | bubble phase of the bed |
| ePh | emulsion phase of the bed |
| EMCD | equi-molar counter-diffusion |
| FB | fluidized bed |
| FBC 2D | (Two-dimensional fluidized-bed combustor) Experimental set-up used here |
| sPh | splash zone of the bed |

furnace as the ones in air-combustion: an inlet O₂ concentration between 25 and 35%_{v/v} is required [2]. Although this first generation reduces the CO₂ emissions, the technology can use higher inlet O₂ concentration in new designs. Despite extensive research [7–10], there is still a number of challenges related to design and building of a second-generation oxy-boiler. Among those are the clarification of the

effect on combustion from changing from an O₂/N₂ atmosphere (air-firing conditions) to O₂/CO₂ (oxy-firing conditions) and the control of the conversion temperature in a smaller reactor with less volume available for heat-exchanger surfaces at high oxygen concentration (> 35%_{v/v}) [11,12]. Pulverized-coal boiler is the main type of boiler in existing power plants, and publications have been focused mainly on

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