



## Study of the high heating rate devolatilization of a pulverized bituminous coal under oxygen-containing atmospheres



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### ABSTRACT

This study deals with the analysis of pulverized coal devolatilization in oxygen-containing atmospheres. To do so, a newly developed flat flame reactor allowing coal jet flames to be stabilized with fuel heating rates of the order of  $10^6 \text{ K s}^{-1}$  has been used to mimic operating conditions close to those met in practical combustors. A high volatile bituminous coal milled in an industrial grinder has been fluidized by carrier gases containing various amounts of oxygen (air and pure oxygen) and then injected in hot gases (temperatures ranging up to  $\sim 1240 \text{ K}$ ) generated by propane/air flat flames. The thermal history of coal particles has been monitored by coupling particle image velocimetry (PIV) and pyrometric measurements to be integrated in constant-rate, single-kinetic-rate, two-competing-rate and DAEM devolatilization models. Devolatilization profiles derived from the analysis of the char collected at different residence times have then been compared with simulated data and a simplified fitting procedure has been implemented to find kinetic parameters leading to reproduce experimental data so as to evaluate the relative influence of the surrounding atmosphere on the devolatilization kinetics. Obtained results confirm that devolatilization is faster and more complete under oxygen enriched environments which has been related to an enhanced combustion of volatiles inducing a rise of the temperature of fuel particles. On the other hand, apparent devolatilization rates appeared to be only indirectly affected by the surrounding atmosphere through the way this one influences the heating of the coal. Furthermore, results obtained in this work tend to indicate the absence of overlapping between devolatilization and char oxidation stages. Measurements of gaseous species released during or formed after the devolatilization process finally confirm that the combustion of volatiles is more complete under oxygen enriched combustion (OEC). More  $\text{CO}_2$  is thus produced while CO concentrations significantly decrease. An enhanced fuel-N conversion has moreover been related to the higher temperatures and oxygen partial pressures met under OEC while  $\text{SO}_2$  concentrations  $\sim 50\%$  higher have been measured due the higher quantities of fuel-sulfur and oxygen available in the medium.

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### 1. Introduction

Coal is currently used to generate around 41% of the electricity in the world with 65% of the global coal demand consumed in the sole power sector in 2010 [1]. Its combustion in coal-fired power plants however results in the generation of carbon dioxide whose emissions account for around 40% of the total quantity of greenhouse gases emitted into the atmosphere [2]. To reduce such emissions,

identified strategies mainly concern the increase of the efficiency of energy production systems and the use of carbon capture and storage (CCS) processes [1].

Among existing CCS techniques, oxycombustion refers to a combustion conducted using a mixture of oxygen and recirculated flue gases (mainly composed of  $\text{CO}_2$ ) [3,4]. An intermediate step between air combustion and full oxycombustion consists in oxygen-enriched combustion (OEC) which is based on the use of air–oxygen mixtures containing elevated  $\text{O}_2$  concentrations ( $>21\%$ ). OEC is actually commonly used in a wide range of industrial applications including mineral production and metal heating or melting for instance [5]. OEC offers a number of advantages in such applications including higher flame temperatures, increased heat releases and boiler thermal efficiency, fuel saving and increased processing rates

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## Nomenclature

$A_{char}$	Dry mass fraction of ash in the char [–]
$A_{coal}$	Dry mass fraction of ash in the raw coal [–]
$dt$	Time step for the devolatilization modeling [s]
$E$	Activation energy [J mol <sup>-1</sup> ]
$E_i$	Activation energy of the $i^{\text{th}}$ reaction [J mol <sup>-1</sup> ]
$E_0$	Central value of the Gaussian distribution of the activation energy [J mol <sup>-1</sup> ]
$F_d$	Devolatilized mass fraction (expressed as a percentage of the original coal) [wt% (DAF)]
$f(E)$	Gaussian distribution function of the activation energy [–]
$f_0$	Volatile fraction contained in the coal (on dry ash free basis) [–]
$k$	Rate constant [s <sup>-1</sup> ]
$k_0$	Pre-exponential factor of the Arrhenius equation [s <sup>-1</sup> ]
$M$	Char residual mass [kg]
$M_{dev}$	Devolatilized mass [kg]
$M_0$	Initial mass of the coal particles [kg]
$M_{inf}$	Mass of volatiles released for an infinite time [kg]
$R$	Ideal gas constant [J mol <sup>-1</sup> K <sup>-1</sup> ]
$T_p$	Coal particle temperature [K]
$T_g$	Temperature of hot gases [K]
$V$	Mass fraction of volatiles released for a given time $t$ (expressed as a percentage of the original coal) [wt% (DAF)]
$V_{inf}$	Mass fraction of volatiles released for an infinite time (expressed as a percentage of the original coal) [wt% (DAF)]
$VM_0$	Volatile matter content of the coal obtained from the proximate analysis [wt% (DAF)]
$X$	Volatile fraction [–]
$\alpha_i$	Mass stoichiometric coefficient of the $i^{\text{th}}$ reaction [–]
$\sigma$	Standard deviation of the activation energy Gaussian distribution [J mol <sup>-1</sup> ]
$\phi$	Equivalence ratio [–]

## Acronyms

C	Coal
CCS	Carbon Capture and Storage
CG	Carrier Gas
DAF	Dry Ash Free
EFR	Entrained Flow Reactor
FF1	Flat Flame 1 (propane/air $\phi = 1.06$ )
FF2	Flat Flame 2 (propane/air $\phi = 1.00$ )
FFR	Flat Flame Reactor
HAB	Height Above the Burner
OEC	Oxygen Enriched Combustion
PC	Pulverized Coal
PIV	Particle Image Velocimetry
$R_c$	Residual char
VM	Volatile Matters
$VM_i$	Volatile Matters issued from the $i^{\text{th}}$ reaction

[5]. Since OEC basically involves removing nitrogen from the oxidizer, the flue gas volumes produced are therefore reduced while their CO<sub>2</sub> fraction increases thus favoring its post-combustion low-cost capture [5,6]. A recirculation of burnt gases (mainly composed of CO<sub>2</sub> and N<sub>2</sub>) can moreover be implemented to increase pollutant concentrations (and ease their treatment) and/or to manage furnace temperatures which in turn leads OEC to be considered as

a retrofit option for traditional pulverized coal (PC) power plants as well as an interesting compromise between conventional air-firing combustion and emerging oxy-fuel combustion systems [6].

This being said, the use of oxygen-enriched atmospheres inevitably affects the ignition and combustion characteristics of pulverized coal particles in addition to pollutant emissions resulting from volatile oxidation and char burnout (see [4,5] for instance and references therein). A thorough understanding of the impact of OEC on the physical-chemical mechanisms involved in the main steps of the thermochemical conversion of pulverized coal is thus needed to analyze the main trends observed during pilot-scale tests as well as to predict scale-up performance through CFD modeling. Devolatilization is the first step in the coal conversion in most common PC boilers. Such a process is consequently of prior importance as it directly influences the subsequent stages. It therefore needs to be well predicted to be able to achieve consistent simulation of PC flames. Different works have thus been undertaken to better understand the influence of the oxygen content of the oxidizer stream on coal devolatilization.

Timothy et al. especially measured average coal particle temperatures between 700 and 800 °C higher when burning lignite and bituminous coals (classified in 38–45 and 90–105 micron size ranges) at 1700 K in a laminar flow furnace using atmospheres containing 100% of oxygen instead of 15% [7]. They also estimated devolatilization times and particle burnout durations between 4 and 5 times lower in 100% O<sub>2</sub> than in air. Finally, predicted devolatilization yields passing from 60 to 80% in 15 and 100% O<sub>2</sub>, respectively, were reported [7]. Similar increases of particle temperatures along with decreases of char and volatile burnout times (depending on the oxygen concentration into the medium) have been reported since then during different drop tube furnace studies [8–10]. As far as devolatilization is concerned more specifically, Murphy and Shaddix reported an apparent enhancement of devolatilization rates of subbituminous and bituminous coals with increasing oxygen concentrations (from 6 mol% to 36 mol%) in a combustion-driven entrained flow reactor (EFR) fueled with particles ground and sieved into a 106–125- $\mu\text{m}$  size fraction [11]. These authors especially related this observation to the combination of two factors including a closer proximity of the volatile flame to the coal particles and the higher temperature of such a flame due to the oxygen-enhanced oxidation of devolatilized matters. Molina and Shaddix [12] then conducted additional experiments in the Sandia's char kinetic EFR with gas temperatures of around 1250 K and concluded that increasing the oxygen concentration (from 21% to 30%) in N<sub>2</sub> accelerates the particle ignition. No clear-cut conclusions were drawn, however, concerning a reduction of the devolatilization time which contrasts somewhat with the results obtained by the same authors when analyzing the ignition and devolatilization characteristics of bituminous and subbituminous coals at a gas temperature of 1700 K and oxygen fractions varying from 12 to 36 vol% [13]. A reduction of around 59% of the devolatilization time was then observed when increasing the O<sub>2</sub> concentration in the proportions indicated above. This trend has been related, as in [11], to an increase of the mass flux of oxygen to the volatile flame which thereby induces enhanced devolatilization rates. Such explanations are consistent with the conclusions issued from a modeling work from Cho et al. who investigated the effects of gas temperature and oxygen concentration, among others, on the volatile matter releases and char burning rates [14]. They demonstrated that the greater the O<sub>2</sub> concentration, the shorter the devolatilization time. By analyzing in detail the results issued from their simulations, Cho et al. indicated that the increase of the devolatilization rate leads to a decrease of the oxygen mass fraction around the fuel particles. These authors argued that the volatiles ejected from the coal particles push away the oxygen and thus generate a depletion zone around the fuel. According to them, this depletion zone is likely to

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