



Full Length Article

Relevance of structure, fragmentation and reactivity of coal to combustion and oxy-combustion



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HIGHLIGHTS

- Coal combustion is influenced by swelling/shrinkage, fragmentation and attrition.
- Coal combustion is influenced by depolymerization, cross-linking and annealing.
- Interplay of chemical and mechanical processes depends on combustion environment.
- Extrapolation of combustion from atmospheric to oxy-fuel conditions is not trivial.

GRAPHICAL ABSTRACT

	Devolatilization	Char combustion	Char gasification
Chemical and microtextural changes	Coal depolymerization, Tar/gas release, Metaplast cross-linking and resolidification	Oxidation by O ₂	Oxidation by H ₂ O, CO ₂ , H ₂
Size and morphological changes	Swelling, Primary fragmentation	Thermal annealing (change of turbostratic carbon structure, ash sintering/melting)	
		Shrinkage, Secondary fragmentation	

Lifetime in a combustor/gasifier →

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ABSTRACT

The nature and extent of the microstructural changes of coal upon oxidation under conditions typical of combustion systems, and the relation between microstructure and reactivity towards oxygen and carbon dioxide have been extensively addressed. The first major changes in morphology, structure and chemical composition of the solid fuel occur in the early instances of combustion/gasification processes, during heat up and devolatilization. At this stage particles can be broken into smaller fragments as a consequence of thermal stress and pressure generated by volatiles release. This phenomenon, regarded to as primary fragmentation, is considered responsible of important changes in the fuel particle size distribution within reactors.

At longer time scales the concurrent heating and heterogeneous reactions may further affect the structure and the reactivity of chars: on the one side the prolonged heat exposure induces thermal annealing with progressive graphitization and loss of reactivity. On the other side, additional phenomena, referred to as secondary fragmentation and attrition by abrasion can occur in parallel with char burning. Eventually, for large extents of internal burning, the increase of particle voidage can induce percolative fragmentation. The distinction between the individual fragmentation processes, just as that between pyrolysis and thermal annealing, is more operational than conceptual and depends on the timescale of the processes and on the nature of the fuel. On the whole, the course of combustion/gasification of a solid fuel is ruled by the competition between heterogeneous reactions of the solid fuel and a number of complex phenomena that modify char structure and reactivity.

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Limited information is available in the literature on the relationship among structure, fragmentation, and reactivity of coal in oxy-combustion conditions. The transferability of the lessons learnt under conventional combustion and gasification conditions to the case of oxy-combustion is here discussed to stress the need of additional investigation on this topic.

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1. Introduction

Oxy-fuel combustion technology received considerable attention in the last decades, being considered the most feasible option for exploitation of coal in the mid-term. An overview on the state of the art of coal oxy-combustion is provided in Refs. [1–4].

In oxy-combustion a mixture of O_2 and recycled flue gas is fed to the combustor to obtain a high- CO_2 flue gas. Therefore, the whole story of a coal particle throughout its lifetime in an oxy-combustor could be significantly changed with respect to conventional air-blown combustion.

The story of a single coal particle throughout its lifetime in a combustor is the result of a complex interplay of phenomena, which affect its chemical, morphological and structural features. These phenomena have been explored in the literature with reference to conventional combustion conditions and should now be considered more in detail also in oxy-combustion processes.

Following the outline of Fig. 1, when a coal particle is injected in a hot reactor, it heats up and rapidly dries (at temperatures of 100–200 °C). Drying is energy intensive and may be accompanied by particle shrinkage. At temperatures between 400 and 700 °C, fuel particles start devolatilizing. Devolatilization consists in the (slightly endothermic) removal of hydrogen- and oxygen-rich volatile matter from the solid fuel matrix, and may be accompanied by particle swelling/shrinkage and/or fragmentation. The volatiles evolved from the fuel particle are further subject to secondary cracking and reforming and may rapidly ignite and burn homogeneously if they come in contact with oxygen. Location of volatiles release and combustion is of great importance for the design and operation of combustors/gasifiers. Drying has typically only a limited influence on fuel pyrolysis, by somewhat delaying the volatiles release [5,6]. However, significant overlapping between drying and pyrolysis has been reported for very coarse and wet fuels [7].

Devolatilization is sometimes accompanied by swelling or shrinking and/or fragmentation of the particles: it is important to account for these phenomena as they change the fuel particle size distribution and, in turn, the characteristic devolatilization time [8].

The char particles left behind by pyrolysis mainly consist of carbon and mineral matter (ash) and have a structure, physical and

chemical properties, and sometimes even a particle size, much different from those of the original fuel particles. The time for char combustion/gasification is typically longer than that for heat up and pyrolysis and, thus, it determines the residence time of the fuel particles in the reactor.

After the early events of pyrolysis and primary fragmentation, char particles can experience over longer time scales, and in parallel with char combustion/gasification, other fragmentation phenomena, referred to as secondary fragmentation and attrition by abrasion. Eventually, for large extents of internal burning, the increase of particle voidage can induce percolative fragmentation, through loss of internal particle wall connectivity.

The following review will address in more detail:

- the chemical reactions that take place along the lifetime of a solid fuel particle in a reactor and their effects and relation with microtextural changes;
- the changes of particles size that a solid fuel particle can experience throughout its lifetime as a consequence of fragmentation phenomena.

A final paragraph will extend these concepts to the case of oxy-combustion. In particular, the relevance of the particle structural modifications in oxy-combustion will be discussed with the aim of highlighting issues which deserve further investigation in the near future.

2. Chemical reactions and microtextural changes

The usual approach for the analysis or prediction of the combustion or gasification of solid fuel particles relies on the assumption that fuel conversion takes place along a purely sequential path consisting of fuel pyrolysis followed by heterogeneous combustion/gasification of its char. According to this framework, heterogeneous combustion/gasification of the raw fuel during pyrolysis would be of limited relevance, as it would be the purely thermally activated modification of the fuel char during the subsequent heterogeneous gasification stage. It has been highlighted how this

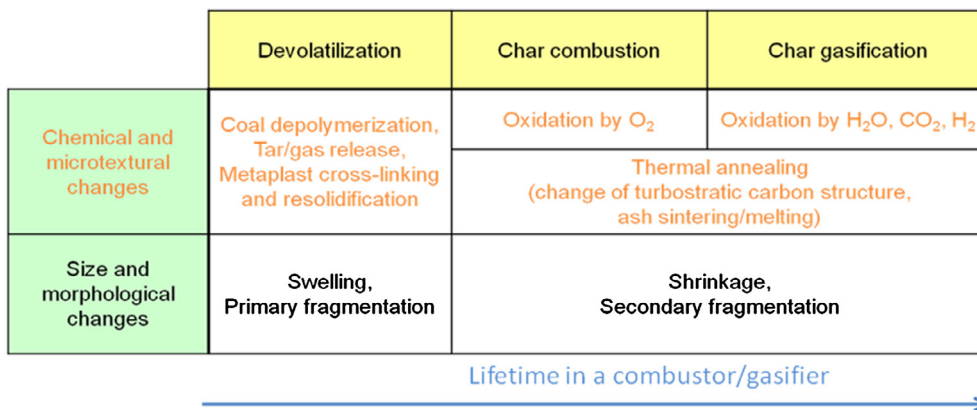


Fig. 1. Outline of processes experienced throughout the lifetime of a coal particle.

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