

Influence of SO₂ on the Fuel Conversion Scheme. Implication for Soot Emissions

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Abstract

This work includes an experimental and modeling study of whether sulfur dioxide (SO₂), typically present in the recirculated flue gas, can inhibit or promote the overall fuel conversion process in a combustion system, with special attention on SO₂ implication for soot emissions and the possible reduction of this pollutant.

Introduction

Conventional combustion processes are typically considered as one of the main responsible of the emission to the atmosphere of different pollutants, with a variety of potential effects on environment and/or human health. Among other important pollutants include: sulfur oxides (SO₂), because of most fossil fuels, biofuels and household waste contain sulfur, and particulate matter (such as soot).

Although technology development and the understanding of combustion and pollutant formation have increased considerably in the last decades, the concern about pollutant formation and emissions continues to be a driving force for the research in combustion chemistry [1]. Flue gas recirculation (FGR) is an interesting technology, both to control NO_x emissions during combustion processes, such as internal combustion engines [2] or combustion systems that incorporate boilers [3], and to control temperature and make-up for the volume of the missing N₂ during oxy-fuel combustion processes [4]. When the FGR technique is applied, reactants are preheated and diluted by recirculated flue gases: mainly CO₂ and water, with minor concentrations of oxygen, CO and other pollutants, such as SO₂, producing thus different effects on combustion process and formation and emission of air pollutants. Thereby, it is of interest to analyze the role of SO₂ on the overall combustion scheme, as a first step towards the further development of flue gas recirculation strategies, and in particular on the emissions of pollutants, such as soot.

The recognized potential hazardous impact of soot on human health has settled the soot chemistry as one of the major research areas in pollutant formation. In general, soot is believed to be formed under fuel-rich conditions at high temperatures [5], through a complex process involving several chemical and physical stages, which is still under research [6].

The presence in the combustion chamber of certain additives or trace species can reduce soot emissions through different ways: i) first, it can inhibit the formation of soot (e.g. by removing the precursor species, the nuclei, or the growth species, or by decelerating the rates of coalescence and coagulation of soot particles), and ii) alternatively, it can remove soot once it has been formed by accelerating its subsequent oxidation, either by stimulating the generation of oxidizing species, such as O or OH, or by direct reaction with the soot [1]. In such a way, SO₂, typically present in the recirculated flue gases, could reduce soot emissions by mutual interactions through any of the soot formation ways, and it could thus be used for soot emission control in a combustion process.

Scope and objectives

In this context, the objective of this study is to identify to what extent the presence of a given compound in the recirculated mixture, in this case SO₂, can affect emissions of a given pollutant, in this case soot, and be used for its control in a combustion process. This research work is considered essential to increase the knowledge on the reaction scheme involving the interactions between SO₂ and hydrocarbons participating in the soot formation process (specifically ethylene), in order to assess the possibility of using flue gas recirculation (FGR) techniques to minimize pollutants emissions, such as soot.

Methodology

This work includes an experimental study on the effect of SO_2 , that can be present when recirculating flue gases, on the overall combustion scheme. Experiments are carried out in a laboratory quartz flow reactor (Figure 1), under well controlled conditions of temperature and inlet reactants concentrations, at atmospheric pressure, in the 975-1475 K temperature range. The concentration of ethylene is kept constant in all the experiments (30000 ppm), whereas the concentration of SO_2 is varied (0-1 %) to address the influence of SO_2 inlet concentration on soot formation. The gases obtained are analyzed and the soot produced is quantified.

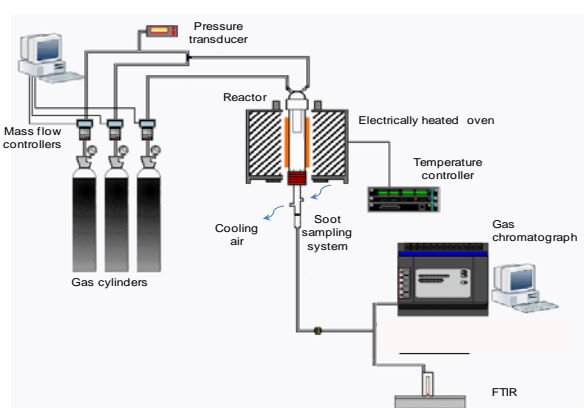


Fig. 1. Scheme of the experimental set-up used for studying the formation of soot.

Additionally, the development of a detailed gas-phase chemical kinetic mechanism for examining the reaction scheme involved in the process is needed. This mechanism should be able to reproduce experimental data in a wide range of operating conditions and, thus, allow analyzing strategies for the minimization of soot precursors in the presence of the different flue gases recirculated, in this case SO_2 .

Conclusions

Main results indicate that the presence of SO_2 in the initial reacting mixture has direct implications on soot formation, achieving different reduction levels as function of the inlet SO_2 concentration. The presence of CO and CO_2 at the reactor outlet points to the fact that oxidation reactions are taking place, and those reactions could come in competition with typical pathways leading to soot formation. The

formation of sulfur compounds such as CS_2 , on the one hand, decreases the amount of carbon available for soot growth and, on the other hand, gives evidence that interactions between sulfur radicals and unburnt hydrocarbons are indeed taking place. The developed model provides, in general, a good description of the gas-phase conversion processes of ethylene, and of the main gaseous products in the presence of SO_2 .

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