



Impact of sour gas composition on ignition delay and burning velocity in air and oxy-fuel combustion



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ABSTRACT

Sour gas is an unconventional fuel consisting mainly of methane (CH₄), carbon dioxide (CO₂), and hydrogen sulfide (H₂S) that constitutes a considerable, currently untapped energy source. However, little is known about its combustion characteristics. In this work, we used our recently assembled and validated detailed chemical reaction mechanism to examine some of the combustion properties of sour gas with different compositions in both conventional air combustion and oxy-fuel combustion, the latter being motivated by application in carbon capture and storage. The calculations suggest that raising the H₂S content in the fuel leads to relatively small changes in the flame temperature and laminar burning velocity, but a considerable reduction in the ignition delay time. At elevated pressures, H₂O diluted oxy-fuel combustion leads to higher burning velocities than CO₂ diluted oxy-fuel combustion or air combustion. Mixed CH₄/H₂S flames exhibit a two-zone structure in which H₂S is oxidized completely to sulfur dioxide (SO₂) while CH₄ is converted to carbon monoxide (CO). Formation of corrosive sulfur trioxide (SO₃) mainly occurs during CO burnout.

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1. Introduction

Sour gas is a special type of natural gas that is currently not being used as a fuel because it contains significant fractions (up to 30% by volume [1]) of hydrogen sulfide (H₂S) and carbon dioxide (CO₂) and so far requires energy-intensive and expensive gas clean-up. Since sour gas resources are significant [2] and there is increasing interest in natural gas as a cleaner alternative to coal for power generation, it is becoming increasingly attractive to develop technologies to overcome this difficulty. One strategy is to use sour gas directly in a gas turbine process employing an oxy-fuel combustion (or oxy-combustion) strategy, possibly combined with enhanced oil recovery. This could help address issues associated with the formation of highly corrosive sulfur trioxide (SO₃) and the low heating value caused by the high CO₂ contents in the fuel [3–5].

However, very little is known about the characteristics of sour gas as a fuel. In particular, it is not known how different

compositions of sour gas affect flame stabilization characteristics or flame structure. There has also been a dearth of experimental information on H₂S oxidation, by itself or as part of a mixture with methane (CH₄). This is the case for conventional air combustion, but certainly even more so for oxy-combustion with its unusual combustion environment consisting of pure oxygen (O₂) and either CO₂ or water (H₂O) as diluents. As a starting point to address this need, we recently assembled and validated a chemical kinetics mechanism for the combustion of H₂S and mixtures of CH₄ of H₂S [3].

The purpose of this paper is therefore to predict some fundamental properties of sour gas combustion under different conditions by means of detailed chemical kinetics calculations, coupled with the corresponding transport properties in the case of premixed flames. The goal is to characterize the influence of varying the fuel composition and to identify possible differences between the different combustion modes (i.e. air combustion vs. CO₂ or H₂O diluted oxy-combustion) based on the observed chemical pathways. We first briefly describe the models used and the parameter ranges considered in the study. Next, we summarize our predictions for the adiabatic flame temperature, ignition delay time, laminar burning velocity, and premixed flame structure of sour gas in air and oxy-fuel combustion using CO₂ or H₂O dilution. Finally, we draw some conclusions for the design of gas turbine combustors burning sour gas.

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2. Modeling

The chemical reaction mechanism for sour gas combustion employed in this study is presented in detail in Ref. [3]. It is a combination of the AramcoMech 1.3 mechanism for the combustion of small hydrocarbons by Metcalfe et al. [6] with an optimized version of the recent mechanism for H₂S oxidation developed by Zhou et al. [7]. The overall mechanism consists of 157 species and 1011 reactions. It has been validated for oxy-fuel combustion of CH₄, air combustion of H₂S, and important interactions between carbon and sulfur species. The scope of the validation was determined by the availability of experimental data [3].

All calculations were conducted in CHEMKIN-PRO [8], using the equilibrium model for the adiabatic flame temperature, the homogeneous reactor model with constant pressure for the ignition delay time, and the flame speed analyzer for the laminar burning velocity and the flame structure calculations. For flame calculations, thermal diffusion of species (the Soret effect) had to be considered.

For the present analysis, sour gas was assumed to be a mixture of CH₄ and H₂S only. The H₂S mole fraction in the fuel was varied between 0% and 30% to account for the range of common sour gas compositions [1]. Carbon dioxide in the fuel was not considered for simplicity, since its influence can be inferred from the calculations for oxy-fuel combustion using CO₂ dilution.

In air combustion, the equivalence ratio ϕ was considered as a variable (design) parameter. For oxy-fuel combustion, the equivalence ratio was fixed at $\phi = 1$, since oxy-fuel systems are likely to operate neither fuel-lean nor fuel-rich to avoid wasting energy for O₂ production or fuel, respectively [9,10]. In this case, the diluent mole fraction can be adjusted instead, since unlike in air combustion the oxidizer and diluent contents are independent.

3. Adiabatic flame temperature

In air combustion, the adiabatic flame temperature can be varied by changing the equivalence ratio. In oxy-combustion, it is controlled by the diluent mole fraction since the equivalence ratio is fixed at $\phi \approx 1$ (cf. Section 2). For simplicity, we only considered atmospheric pressure without preheating ($p = 1$ atm, $T_{in} = 300$ K). The same qualitative trends also apply at elevated pressure and temperature.

Regardless of the combustion mode, the flame temperature decreases by about 30–50 K when raising the H₂S content in the fuel from 0% to 30% (see Fig. 1). This is caused by the smaller lower heating value (LHV) of H₂S, which according to calculations using the same thermochemical data is only 65% of the heating value of CH₄ (LHV_{H₂S} = 518 kJ/mol as compared to LHV_{CH₄} = 803 kJ/mol).

In the case of oxy-fuel combustion, the adiabatic flame temperature is higher when using H₂O as a diluent as compared to CO₂. This difference can be explained with the higher isobaric heat capacity (c_p) of CO₂ (e.g., $c_{p,CO_2} = 58.4$ J/molK at $T = 1500$ K as compared to $c_{p,H_2O} = 47.1$ J/molK).

4. Ignition delay time

The ignition delay time is a common metric for describing the oxidation characteristics of a fuel and can give some indications of flame stabilization behavior. If the fuel is to be used in a gas turbine employing premixed combustion, the ignition delay time is important in determining the possibility of autoignition that can damage the equipment [11].

For the present analysis, we defined the ignition delay time using the temperature inflection point (see Fig. 2). However, the results presented here are not sensitive to the exact definition of

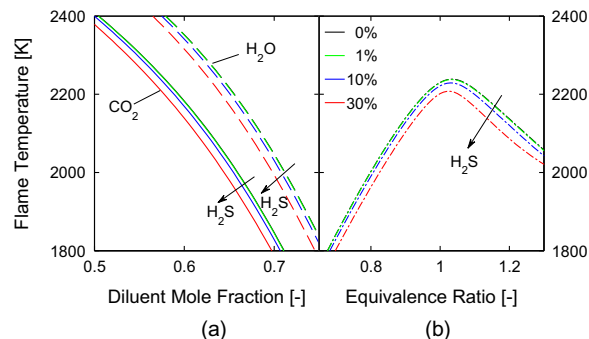


Fig. 1. The adiabatic flame temperature of sour gas decreases with increasing H₂S content in the fuel: (a) Oxy-fuel combustion with CO₂ dilution (solid lines) and H₂O dilution (dashed lines) and (b) air combustion.

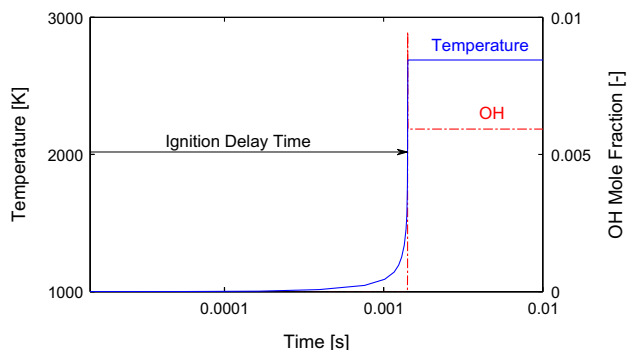


Fig. 2. Definition of the ignition delay time by the temperature inflection point. The OH mole fraction is shown for comparison. Sour gas (30% H₂S, 70% CH₄) in air at $\phi = 1$, $p = 40$ atm, $T_0 = 1000$ K.

the ignition delay time. For all cases, we considered a constant pressure of $p = 40$ atm and an initial temperature of $T_0 = 800$ – 1200 K. We decided to use constant pressure rather than constant volume simulations in order to account for the fact that in gas turbine combustors, pressure typically stays constant within a few percent [11]. The values of pressure and temperature are chosen to be representative of the inlet conditions of gas turbine combustors, but also close enough to the conditions for which the H₂S kinetics are validated [3]. However, preliminary calculations at different pressures between $p = 1$ atm and $p = 80$ atm revealed the same qualitative trends.

4.1. Impact of fuel composition

The ignition delay time of sour gas in air decreases with increasing H₂S content in the fuel, indicating that the low and intermediate temperature kinetics of the mixture get faster (see Fig. 3). At low temperature ($T_0 \approx 800$ K), addition of only 1% H₂S already lead to a substantial decrease (–65%) in the ignition delay time as compared to pure CH₄. Higher concentrations of H₂S further decrease the ignition delay time only slightly (up to –83% for pure H₂S as compared to pure CH₄). At higher temperature ($T_0 \approx 1200$ K), however, the promoting effect of small concentrations of H₂S is less pronounced while higher concentrations have a stronger promoting effect than at lower temperature. The same qualitative behavior was also observed for oxy-fuel combustion using CO₂ or H₂O dilution (not shown here).

Addition of H₂S has a significant impact on the radical pool. Compared to pure CH₄, small amounts of H₂S lead to a much faster buildup of H, OH, HO₂, and in particular O (see Fig. 4, note the different timescales in the two subfigures). It also enhances the

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