Premixed methane oxycombustion in nitrogen and carbon dioxide atmospheres: measurement of operating limits, flame location and emissions. Proceedings of the Combustion Institute

R. Marsh*, J. Runyon, A. Giles, S. Morris, D. Pugh, A. Valera-Medina, P. Bowen

Cardiff University, School of Engineering, Cardiff CF24 3AA, United Kingdom

Abstract

The influence of O_2 concentration on a premixed swirling flame was investigated for changes in discrete levels of CO_2 and N_2 dilution, with application to carbon capture in gas turbine systems. Chemical kinetic models were utilised to support and analyse generic burner experiments at 37.5 kW. O_2 mole fraction in the oxygen-diluent stream was varied between 0.21 and 0.70 and careful measurement of velocity field data using particle image velocimetry, representative heat release (OH^* chemiluminescence) and flame location (OH planar laser-induced fluorescence) was undertaken. Results show that under lean N_2-diluted operating conditions there is a minor change in burner operation for all O_2 concentrations considered. CO_2 dilution has a far more substantial impact than N_2 on flame location, heat release and operational response, which is attributed to the fundamental differences in thermodynamic and transport properties between the two gases. This also resulted in increased CO concentrations sampled from the exhaust stream with a rise in diluent CO_2, which is attributed to lower flame temperatures as opposed to thermal dissociation, whilst increased N_2 dilution resulted in increasing NO_X emissions.
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1. Introduction

The aim of this research is to analyse the characteristics of CO_2 and N_2 diluted swirl-stabilised combustion of O_2 and CH_4 mixtures; principally to show how flame location changes in relation to O_2 mole fraction and equivalence ratio, in con-

* Corresponding author.
  E-mail addresses: marshr@cardiff.ac.uk, marshrichard@hotmail.com (R. Marsh).

Nomenclature

Da	Damköhler number
l_0	integral length scale
u'	axial velocity
α	thermal diffusivity
S_L	laminar burning velocity
ρ	density
c_P	molar heat capacity at constant pressure
λ	thermal conductivity
μ	dynamic viscosity
β	molar oxygen concentration in oxidant mixture
ϕ	equivalence ratio
S_g	geometric swirl number
Q	molar flow rate
$D_{3,2}$	Sauter mean diameter

Table 1
Properties of oxygen and diluents at 1000 K and 0.1 MPa.

Substance	O_2	N_2	CO_2
ρ (kg/m^3)	0.385	0.337	0.529
c_P (J/mol.K)	34.9	32.7	54.3
λ (10^3 W/m.K)	79.7	66.0	70.6
μ (10^6 Pa.s)	49.1	41.6	41.3

1.1. Chemical timescales

Robust understanding of the phenomena governing premixed turbulent combustion involves consideration of chemical and flow timescales within the reaction zone. In the case of methane oxycombustion with variable diluents, the interaction between chemical and flow properties must be measured. These are described by consideration of the flame Damköhler number, defined in Eq. (1) as:

$$Da = \frac{(l_0/u'_{RMS})}{(\alpha/S_L^2)} = \frac{\text{flow time scale}}{\text{chemical time scale}} \quad (1)$$

Different velocity, length and chemical timescales lead to a variety of extinction conditions, thus affecting blowoff conditions [17].

Diluents affect the flame in at least four ways: (1) mixture heat capacity, (2) transport properties, (3) chemical kinetics, (4) radiative heat transfer [15]. A reduction in burning velocity is noted when N_2 is replaced by CO_2—primarily attributed to the higher heat capacity of CO_2 [18]. Table 1 shows physical properties for each substance [19].

2. Methodology

2.1. Mixture convention

Oxygen mole fraction (β) is defined as the molar ratio of oxygen in the oxygen/diluent mixture, Eq. (2), in the case of the N_2 system (where for air $\beta = 0.21$) [4]:

$$\beta = \frac{Q_{O_2}}{Q_{O_2} + Q_{N_2}} \quad (2)$$

As β is variable in the oxidant mixture, care must be taken to identify both equivalence ratio (ϕ) and β when presenting results. Since conditions studied herein were designed to evaluate the behaviour of variable O_2 content on premixed swirl burners, all data were collected in the region $\phi < 1$ and $\beta > 0.21$.

2.2. Chemical kinetics simulation

The PREMIX coded laminar flame speed calculator was utilised in CHEMKIN-PRO [20,21] to model the reactant mixtures for ambient conditions of temperature (293 K) and pressure (0.1 MPa)

junction with the local flow field of the swirl zone. Limited combustion research has been published in this field, where premixed or partially premixed natural gas has been burnt in O_2–CO_2 mixtures [1–10]. Recent studies on gas turbine burners used exhaust gas recirculation levels up to 30%, and research with oxygen-enhanced natural gas burners has considered molar O_2 fractions up to 40% [11,12]. Other work demonstrated stability limits of tubular burners with various O_2 and CO_2 concentrations, proving O_2–CH_4 flames can be balanced with CO_2 as a combustion moderator [13]. Studies on flame front locations demonstrated flame shape is influenced by O_2 fraction, but weakly dependent on equivalence ratio [2]. When comparing diluents, limited studies show that at constant mass flux, CO_2 diluted flames are stabilised at higher O_2 concentrations compared to N_2. Such reaction kinetics have been studied with CHEMKIN to show differences in laminar burning velocity of O_2–CH_4 mixtures with N_2 and CO_2 diluents; it should be noted also that CO_2 contributes to soot oxidization and affects the rate of many chemical reactions compared with N_2. Under stoichiometric conditions, a pure O_2–CH_4 flame exhibits flame speeds around 10 times greater than the air–CH_4 equivalent [14]. Moreover, premixed O_2–CH_4 swirl stabilised burners, also diluted with N_2 and CO_2, demonstrated dependence of strain rate through chemical kinetics and flow residence times on flame blowoff [15]. A rapidly mixed tubular burner has shown how oxygen concentration with CO_2 and N_2 diluents affected stability limits, utilising Damköhler number to describe the flow field in relation to mixing and chemical residence times [16]. This work also demonstrated that CO_2 dissociation in oxygen–methane flames is not significant at temperatures below 2400 K.