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Shock tube ignition delay times and methane time-histories measurements during excess CO_2 diluted oxy-methane combustion

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ABSTRACT

The combustion of methane in air results in large amounts of CO_2 and NO_X emissions. In order to reduce the NO_X emissions, one possible solution is the oxy-methane combustion with large CO_2 dilution so that the combustion products can be reduced mainly to CO_2 and H_2O. However, there are very few studies on the chemical kinetics of oxy-methane combustion in a CO_2 diluted environment. In this study, methane time-histories, CH* emission profiles, and pressure time-histories measurements were conducted behind reflected shock waves to gain insight into the effects of CO_2 dilution of the gas mixtures on the ignition of methane. The measurements were carried out for mixtures of CH_4, CO_2 and O_2 in argon bath gas at temperatures of 1577–2144 K, pressures of 0.53–4.4 atm, equivalence ratios (Φ) of 0.5, 1, and 2, and CO_2 mole fractions (X_{CO2}) of 0, 30, and 60%. The laser absorption measurements were conducted using a continuous wave distributed feedback interband cascade laser (DFB ICL) centered at 3403.4 nm. The results showed the decrease of activation energy and the increase of ignition delay time as the amount of CO_2 dilution was increased. However, the changes were minor and within the experimental uncertainties of the measurements. Also, the results were compared to the predictions of two different natural gas mechanisms: GRI 3.0 and AramcoMech 1.3 mechanisms. In general the predictions were reasonable when compared to the experimental data; however, there were discrepancies at some conditions. Three different influences of CO_2 addition to the argon bath gas in regards to chemistry, collision efficiencies, and heat capacities were examined. In addition, the present study included experimentally obtained correlations for absorption cross sections of methane for its P(8) line in the v_3 band in argon bath gas with and without carbon-dioxide dilutions at temperatures between $1200 < T < 2000$ K and pressures between $0.7 < P < 1.2$ atm.

1. Introduction

Energy consumption has increased dramatically as the world advances and becomes more industrialized. Over the next twenty five years, the U.S. Department of Energy expects the energy demand to increase by 29% with almost all of the new energy from natural gas [1]. A problem is that current methods for the combustion of natural gas (e.g., gas turbines) result in large amounts of CO_2 and NO_X emissions. In order to reduce the greenhouse gases, one possible solution is the oxy-methane combustion with large CO_2 dilution. By using pure oxygen instead of air, the resulting products can be reduced to mainly CO_2 and H_2O. H_2O can be condensed out and remaining CO_2 can then be captured and returned to the power cycle or stored underground. The concern is the difference in methane oxidation in air vs CO_2 mixtures. It has been shown that the reactions behave differently as the properties of nitrogen and carbon dioxide differ [2] in terms of participation in combustion reactions directly or as a third-body collision partner. As a result, more analysis of oxy-methane combustion with high CO_2 addition needs to be conducted.

There are some studies of CO_2 diluted oxy-methane combustion in the literature. Heil et al. investigated the methane burning rates for flameless combustion and compared the results to nitrogen diluted mixtures [3]. Di Benedetto et al. and Liu et al. looked at the chemical effects (flammability and burning velocity) of methane combustion in CO_2 versus N_2 [4,5]. The laminar flame speeds have also been studied for various conditions [6–9]. In addition, Vasu et al. examined the effect of CO_2 dilution on the ignition delay times of syngas mixtures of hydrogen and carbon monoxide [10]. However, there are very few studies in the literature that probed the effects of excess CO_2 dilution on the ignition delay times of methane. Holton et al. conducted ignition delay time measurements of natural gas blends, including methane and ethane mixtures, with small amounts of CO_2 addition (5 and 10%) [11]. They found out that methane and ethane blends at $\Phi = 0.5$ and $T = 1137$ K diluted with 5% CO_2 increased the ignition delay time by only 2%, whereas 10% CO_2 addition to the same mixture resulted in longer times by 46%. This increase was attributed to

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Table 1
Ignition delay time simulation predictions at 1600 K and 1 atm.

	X_{AR}	X_{N2}	X_{CH4}	X_{O2}	X_{CO2}	$\tau_{AramcoMech\ 1.3}$ [μs]	$\tau_{GRI\ 3.0}$ [μs]	$\Delta\tau_{dif}$
Ar bath	0.895	0	0.035	0.07	0	1495.5	1090.1	405.5
	0.595	0			0.3	1788.3	1325.8	462.5
	0.295	0			0.6	2024.9	1547.4	477.5
N_2 bath	0	0.895	0.035	0.07	0	1665.8	1164.8	501.0
	0	0.595			0.3	1865.8	1362.5	503.3
	0	0.295			0.6	2059.4	1560.0	499.4

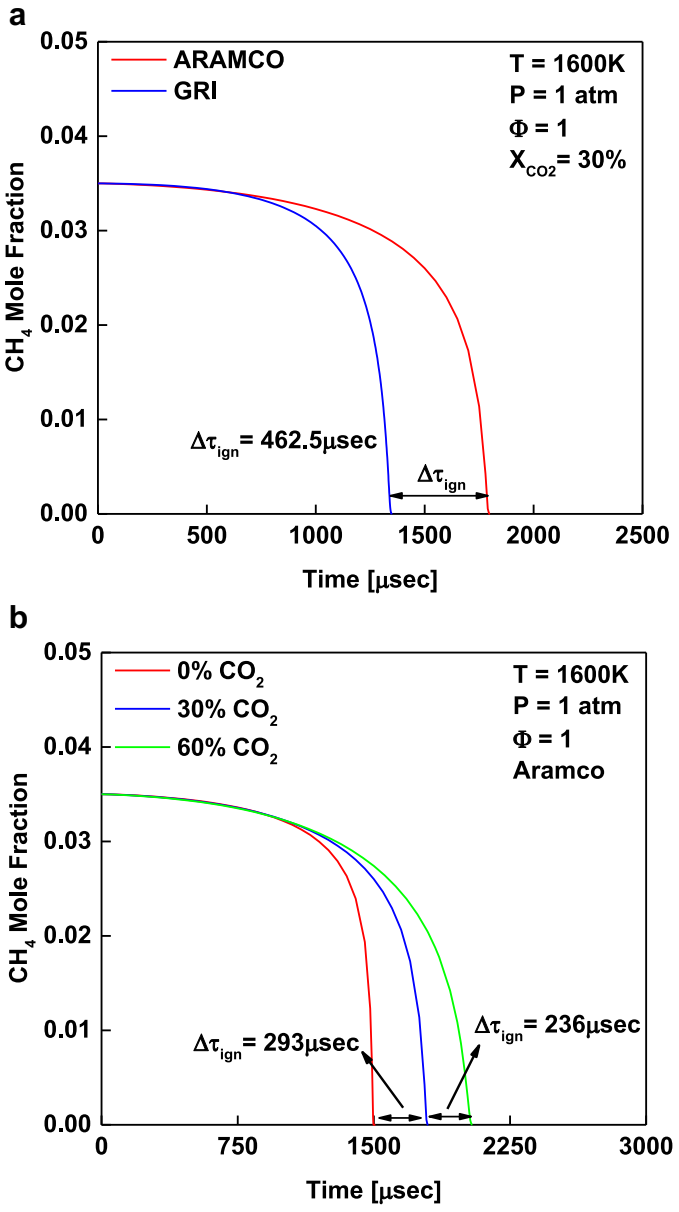

Fig. 1. (a) Comparison of methane time-history predictions obtained from GRI 3.0 and AramcoMech 1.3 mechanisms for the stoichiometric combustion of 3.5% CH_4 in 30% CO_2 in argon bath gas at 1600 K and 1 atm; (b) methane time-histories during its ignition when the bath gas contains different percentages of CO_2 ranging from 0 up to 60% according to the AramcoMech 1.3 mechanism. Note: The reader is referred to the online version of this article for better color clarity for all figures.

the third-body collision efficiencies of CO_2 being an order of magnitude greater than those of N_2. However, they suggested carrying out further experiments in order to better quantify the effect of CO_2 addition on the ignition delay time.

Figure 1(a) gives the comparison of methane time-history predictions of two different reaction mechanisms; namely the GRI 3.0 and the AramcoMech 1.3 [12,13], for stoichiometric combustion of 3.5% CH_4 in argon bath gas diluted with 30% CO_2 at 1600 K and 1 atm. The results were obtained using the constant- volume, internal energy (constant-U,V) assumption with the CHEMKIN PRO tool [14]. The discrepancy in the ignition delay time between the two mechanisms turned out to be $\Delta\tau_{ign} = 462.5$ μs. Figure 1(b) shows CH_4 time-histories during its ignition when the gas mixture contains different mole fractions of CO_2 ranging from 0 up to 60% according to the simulations done with the AramcoMech 1.3 mechanism. The differences in the ignition delay times were $\Delta\tau_{ign} = 293$ and 236 μec when X_{CO2} was increased from 0 to 0.3 and 0.3 to 0.6, respectively. These variations in the predictions of two chemical mechanisms with the addition of CO_2 necessitate conducting validation experiments on CH_4 ignition with CO_2 dilution.

Although not shown in Fig. 1(a) and (b), the discrepancies in the predicted ignition delay times between the two mechanisms were noticed in N_2 and Ar bath gas even without any CO_2 dilution. These ignition delay time simulations at different bath gasses and CO_2 dilutions at 1600 K and 1 atm are summarized in Table 1. It can be seen from the table that as the CO_2 dilution was increased from 0 to 60%, the differences ($\Delta\tau_{dif}$) between the two mechanisms raised from 405.5 to 477.5 μs in argon bath. However, the difference between the two mechanisms remained the same (499.4 μs $< \Delta\tau_{dif} <$ 503.3 μs) when nitrogen was used as the bath gas. Also, differences in the ignition delay times within the mechanisms themselves were seen as the CO_2 dilution was raised. This was already exemplified in Fig. 1(b), but further detailed in Table 1. As the CO_2 amount was increased, it was observed that the changes in the ignition delay time were more significant when the bath gas included argon (e.g. an increase from 1495.5 to 2024.9 μs for AramcoMech 1.3 mechanism) than nitrogen (e.g. an increase from 1665.8 to 2059.4 μs for AramcoMech 1.3 mechanism).

In this study we provided ignition delay time measurements for mixtures of CH_4, CO_2, and O_2 in argon bath gas at temperatures of 1577–2144 K, pressures of 0.53–4.4 atm, equivalence ratios (Φ) of 0.5, 1, and 2, and CO_2 mole fractions (X_{CO2}) of 0, 0.3, and 0.6. The measurements were done by utilizing a recently built shock tube facility at the University of Central Florida (UCF) in the reflected shock region. Experimental data were compared to the predictions of two different kinetic models: GRI 3.0 and AramcoMech 1.3 mechanisms [12,13]. The ignition delay time measurements showed the influence of CO_2 dilution on the oxidation of methane. In addition, we built a laser absorption diagnostic for measuring CH_4 time-histories behind the reflected shock waves using a continuous wave distributed feedback interband cascade laser (DFB ICL) centered at 3403.4 nm. The present study included experimentally obtained correlations for absorption cross sections of CH_4 for its P(8) line in the ν_3 band (λ = 3403.4 nm) in argon bath gas with ($X_{CO2} = 0.3$) and without ($X_{CO2} = 0.0$) CO_2 dilutions at temperatures of 1200 $< T <$ 2000 K and pressures of 0.7 $< P <$ 1.2 atm. CH_4 time-histories during stoichiometric ignition of CH_4 with and without CO_2 dilution around 1 atm were also obtained through the aforementioned absorption cross section correlations. To the best of our knowledge, the current study provides