Measurement Of CO_2 Emission And Stable Gas Species Concentrations Within A Natural Gas Diffusion Flame

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Abstract

Measurements of temperature and major stable gas compositional change within the axis of a natural gas diffusion flame has been carried out and reported in this work. The mole fractions of CO_2, CH_4, C_2H_6, C_3H_8, C_4H_{10}, C_5H_{12} and others were measured using an isokinetic sampling probe insert and gas chromatophy, at several axial locations within the flame, particularly with respect to axial distances from the burner rim. Two flames of equivalence ratios 0.35 and 1.70 were considered. General observation on temperature relationship with C_1, C_2, C_3, C_4, and C_5, for equivalence ratio of 0.35 flames can be summarized to be inversely proportional. For all the cases considered, increase in species concentration always corresponds with decrease of temperature. An important fact to note occurred when all the species (C_1, C_2 and C_3) concentrations for $\Phi = 1.70$ dropped to almost zero at the flame tip of 30cm, which is the flame height. At this point it is believed that the species were fully consumed and this point could be taken to be stoichiometric. This work provides comprehensive experimental data on the characteristics and structure of natural gas diffusion flame which are essential for computational studies on these flames.

Introduction

The use of natural gas (one of the fossil fuels) in the world today is fast expanding largely because of its relative abundance and also because of its low level emissions of nitrogen, carbondioxide and unburned hydrocarbons depending on the condition of usage and type. Other advantages include its wider availability, usage and low cost of the fuel. Its major disadvantages lie on the need for ignition enhancement in vehicles. This is highly necessary in that its auto ignition temperature condition is within the neighbourhood of 1200K, [13].

Natural gas being a mixture of methane, ethane, propane, butane, pentane, hexane, carbon dioxide and nitrogen in various proportions, (the proportion and composition depends on the reservoir formation) is finding enormous application both in automotive systems and in power plant systems. It has been vastly applied in the following areas of engineering endeavors; gas turbines, gas engines, industrial boilers and vehicles, while domestic or residential use has been presently developed [12]. No doubt about it, on the extent natural gas usage will go, its number 1. To natural gas executives of three 'supermajors' believe natural gas is ready to overtake crude oil as the preferred fuel, for virtually all combustion related facilities, [4].

Gaseous emissions, as features of fossil fuel combustion, influence to a large extent, the process efficiency in the user combustion system. It has been reported by several researchers that it has some strong negative impact on the process kinetics and on the environment, including human health (some gaseous emissions are medically known to be caseinogens), [8,1]. Several researches, like the works of Kennedy et al [7], Musick et al [10], Tao et al [13], Gore and Zhan [3], Bernstein, et al [2], and Miller et al [9] have presented detailed experimental results on gaseous emission concentrations with respect to some coordinates within and outside the contour of the resulting flames. Also, Joao Cassiano et al [5] and Keller et al [6] worked extensively on NOx and CO emissions from industrial combustions operated under different modes. The aim of this present research is to study the combustion characteristics of Nigerian natural gas flame, gaseous product species and compositional changes in a simulated locally designed combustion test-rig. It provides information on the quantitative dependence of the production of some gaseous species on the temperature and equivalence ratios of 0.35 and 1.70.

Experimental Procedure

An over-ventilated and under-ventilated flame of natural gas of equivalence ratios 0.35 and 1.70 was established on a co-annular burner using an in-house developed experimental test rig designed for diffusion flame studies. This burner consisted of a 16mm diameter steel fuel tube (where the natural gas is injected upwardly) and surrounded by a 109mm diameter air annulus. The resulting flame was shielded by square shaped sheet (casing) to protect it from air movement in the laboratory. There exist probe insert holes on one side of this casing for Isokinetic sampling. Also, there exist a window on the other side of the casing made of tough 4.5mm thick glass material, which provides optical access to the chamber for photographic shots and for visual inspection of the flame. The resulting diffusion flame temperature was measured with a K – Type Thermocouple.

Temperature Profile Measurement

A k – type (Chromel - Alumel) thermocouple, which has been calibrated with Fluke 5502 Multi-product, Multi-function calibrator, was used to measure axial and

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radial temperature profiles within the flame. The thermocouple with a well-designed attachment was inserted through the sample pots (fifteen in number) made at the right hand side of the chamber. The temperature profile was measured for two different diffusion flames of equivalent ratios 0.35 and 1.70. That is, first flame is under a lean condition while the second flame is a rich mixture. Details of the operating conditions adopted in this work are shown in tables 1 and 2, while figure 1 shows the burner pipes. At normal environmental condition of temperature 25^0 C (298K) and pressure 1.013bar, the fuel and the oxidant (air) possess the following properties:

Natural Gas: Density $\rho_{ng} = 0.874$ (kg/m^3), $\mu_{ng} = 1.45 \times 10^{-5}$ kg/ms, $D_{ngn} = 0.016$m, $A_{ngn} = 2.0 \times 10^{-4}$ m^2. Molecular weight, $M_{ng} = 18.48$ g/mol.

Oxidizer Gas (Air): Density $\rho_a = 1.177$ (kg/m^3), $\mu_a = 1.846 \times 10^{-5}$ kg/ms, $D_{a,n} = 0.109$m, $A_{an} = 9.33 \times 10^{-3}$ m^2, Prandtl number (Pr) = 0.707.

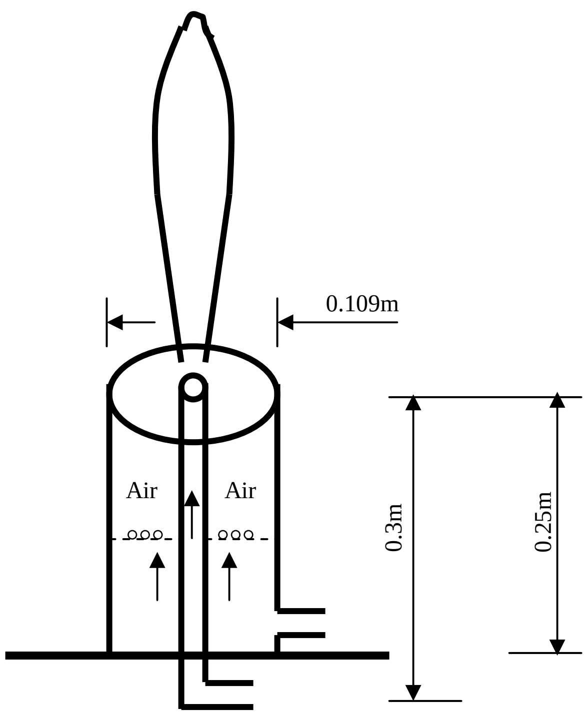

Fig. 1: Details of the fuel Nozzle and Burner dimensions.

Table 1: Operating Conditions of the fuel (Natural Gas)

S/N	Z	Φ	m_{ng} (kg/sec) x 10^{-5}	Q_{ng} (m^3/sec) X 10^{-5}	Q_{ng} (l/m)	V_{ng} (m/s)	Re_{ng}
1	0.02	0.35	0.73	0.83	0.50	0.042	41.0
2	0.09	1.70	2.91	3.33	2.00	0.167	161.0

Table 2: Operating Conditions of the Oxidizer (air)

S/N	Z	Φ	m_a (kg/sec) x 10^{-4}	Q_a (m^3/sec) x 10^{-4}	Q_a (l/m)	V_a (m/s) x 10^{-2}	Re_a
1	0.02	0.35	2.94	2.5	15	2.68	186
2	0.09	1.70	2.94	2.5	15	2.68	186

Gas Sampling Procedure

Isokinetic sampling technique adopted by Gore and Zhan (1996) is partly employed in this research. Isokinetic sampling technique is a process of drawing a sample from a moving stream of gas (in this case, natural gas + air combustion flame) at the same velocity as that of the free stream. It can be water or air cooled depending on choice and condition of operation. The water – cooled sampling line has the arrangement shown in Ofodu et al [11]. To take a sample, the probe is moved quickly to the desired axial positions (0,3,6,9,12,15,18,21,24,27,30) cm. There exist two sets of bottle; the sampling bottle A, (which should exist at a reduced pressure than 1atm) and the purge bottle B, (containing either helium or nitrogen gas existing at over – pressure for purge flow). Once the sample is to be taken, as the probe is moved to the desired position, a small purge of helium or nitrogen gas was admitted. At this point, the sampling bottle was isolated. Once the probe is at the appropriate location, the purge flow valve is closed and the bulb isolation valve (leading to the sample bottle A) is opened. When the sampling end, at this time, the bulb isolation valve (for sampling bulb) is closed, gas samples approximately 10ml volume is removed with a syringe for chromatographic analysis of the gaseous species.

Gas Chromatography

The gas sampled in the syringe was analyzed using **Agilent 6890N Gas Chromatograph** equipped with two switching valves, a heated inlet, two columns housed in an oven and two detectors. The sample first passes over a heated inlet through the switching valves that injects the samples into the two columns. The packed **Chromosorb Column** separates the non-hydrocarbons (O_2, N_2, CO_2) and the **Plots-Alumina Capillary Column** separates the hydrocarbons (C_1 to C_{10}). The separated components from the packed column are detected by a **thermal conductivity detector** while that of the capillary column was detected by **Flame Ionization Detector**. The peak areas of the **Chromatogram** are interpreted using **CHEM-STATION** software by comparing peak areas and retention time of the sample with those obtained on a reference standard mixture of pure hydrocarbon. Subsequently, an in-house simulator performed the chromatographic analysis, which finally presents the results in mole fractions of the species present.

Results and Discussion

The plots of the gaseous species and the compositional changes are presented with respect to axial distance and equivalent ratio. Also, the density (g/cm^3) is shown for equivalence ratios of 0.35 and 1.70. Axial temperature plots for equivalence ratio of 0.35 (for natural gas) are shown in figure 2 in conjunction with those obtained from Musick et al [10] for methane flame.

Gaseous Species Profile For Φ = 0.35

XCO_2 And Gas Density Axial Profiles

It is evident that the concentration of carbondioxide (XCO_2) decreases gradually in mole fraction from the value of 0.312 at the burner rim to 0.0028 at about 12cm at the rate of 0.24% per cm distance. On reaching at the position of 18cm it increased to the value of 0.0093. The 18cm position corresponds to the point of stoichiometric mixture and also matches with the point of flame tip (the height of flame is 18cm). Within these range of positions considered, the temperature behaved in an inverse manner, that is; increases whenever CO_2 is decreasing and subsequently decreases whenever CO_2 is increasing. It is evident from the graph of figure 3 that both actions occur at the same rate. Also, the flame gas density variation along the axis of the flame is graphically shown in figure 3. The density with the value of 0.00106 g/cm^3 at the burner rim decreases continuously to the value of 0.0009 g/cm^3 at 12cm and subsequently increased to 0.00144 g/cm^3 at the flame tip of 18cm.

XC_1, XC_2 And XC_3 Axial Compositional Change

Figure 4 shows the graphical plots of the mole fractions of C_1, C_2 and C_3 along side with temperatures with respect to centerline distance. It is observed that for all the positions considered (1,6,12 and 18cm), that:

$C_1 > C_2 > C_3$ in mole fractions and all maintained the same rate of decrease from the values of 0.781, 0.0454 and 0.0340 (at the burner rim) to 0.5419, 0.0265 and 0.0162 (at axial position of 6cm) and subsequently increased to 0.5979, 0.0421 and 0.0229 (at axial position of 12cm) and finally decreased to same value at position 18cm.

XiC_4, XnC_4, XiC_5, and XnC_5 Axial Profile

The mole fractions of isomers and normal Butane (iC_4 and nC_4) and pentane (iC_5 and nC_5) respectively with respect to axial positions within the flame as shown in figures 5 and 6, all behaved in the same manner. Both plots decrease from their values at the burner rim (0.0147 and 0.0195) for butane to lower values at 6cm and 12cm axial positions and subsequently increased to about same values as that at the burner mouth for butane and to 0.0261, 0.0081 for pentane isomers and normal respectively at the flame tip.

Gaseous Species Profile For Φ = 1.70

XCO_2 And Gas Density Axial Profile

Concentrations in mole fraction of CO_2, stream gas density and position temperature with respect to distances at the flame centerline are shown in the graph of figure 7. This flame, being of equivalence ratio of 1.70 is rich. It is noted that CO_2 decreased from 0.1192 to zero and maintains this until after axial position of 12cm it increases to 0.2598 at 18cm axial position. Subsequently, it decreases to a lower value of which it maintains to the flame tip. The axial density variation was just as equivalence ratio of 0.35.

XC_1, XC_2 And XC_3 Axial Compositional Change

Figure 8, shows the axial distribution of some stable gaseous species within a natural gas diffusion flame operating at a rich condition of Φ1.70 equivalence ratio. While C_1 was decreasing in mole fraction, C_2 and C_3 were increasing gradually as the stream of flame flows from the burner rim (at x = 0) to axial position of 12cm. After this point, C_1 made gradual increase while C_2 and C_3 made a sharp decrease and simultaneously increased to maximum at a position x = 24cm. An important fact to note occurred when all the species (C_1, C_2 and C_3) present in this condition dropped to same value approximately at the flame tip of 30cm, which marks the flame height. At this point it is believed that they were fully consumed and that gives the striochiometric region.

XiC_4, XnC_4, XiC_5, and XnC_5 Axial Profile

Figures 9 and 10 show the mole fractions of isomeric and normal butane and pentane respectively, within a natural gas diffusion flame of 1.70 equivalence ratio. The behaviour of these isomers of C_4 and C_5 showed same characteristics of gradual increase and decrease within the neighbourhood of 0cm to 18cm axial positions. At 24cm axial position they all sharply increased to maximum and decreased to a lower value at 30cm.

Comparative Study of Species Variations at Lean and Rich Conditions

This section presents, the axial variation in the concentrations of XC_1, XC_2 XC_3 and XCO_2 for both lean and rich conditions of the flame. Figures 11, 12, 13 and 14 show their comparative plots. In figure 11, both flames exhibited same mole fraction at the burner rim (XC_1 = 0.78) and both decrease (with that of Φ = 1.70 being at a faster rate than Φ = 0.35). This behaviour continues until a crossover occurred at the axial position of about 15cm. From figure 12, both flames recorded same value of C_2 at the burner rim. For other axial positions, C_2 is higher for 1.70 equivalence ratio. The C_3 mole fraction at equivalence ratio of 0.35 (figure 13) at the burner rim was higher than the other flame. At about 2cm a crossover occurred, which kept C_3 at Φ1.70 higher for all axial positions. Figures 14 and 15 are for CO_2 and gas density respectively.

Conclusion

This paper presents the results of measurements of temperature and major stable species compositional changes within the vertical axis of a natural gas diffusion flame. The study was carried out with a combustion facility designed using the technique, adopted from Gore and Zhan (1996). The mole fractions of CO_2, CH_4, C_2H_6, C_3H_8, C_4H_{10}, C_5H_{12} and others were measured using isokinetic sampling probe insert and gas chromatography, at several axial locations within the flame. Two flames of equivalence ratios 0.35 and 1.70 were considered. This provides compositional and concentration changes to be measured to determine the effect of equivalence ratio on the species concentration levels in these flames.

Acknowledgement

The authors acknowledge the help and valuable discussion and inputs made by Engr. Prof. D.P.S Abam, Department of Mechanical Engineering, University of Port Harcourt and Engr. Prof. K.D.H Bob-Manuel of RSUST, Nigeria. The overwhelming assistance provided by Dr. Michael Balthasar, University of Cambridge, and Prof. Ian, Kennedy of University of California are all well acknowledged.

REFERENCES

1 Bai, X.S., Balthasar, M., Mauss, F. and Fuchs, L., Detailed Soot Modeling in Turbulent Jet Diffusion Flames, *Twenty-Seventh Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, 1623-1630 (1998).

2 Bernstein J.S, Fein Asa, Choi J.B, Cool T.A, Sausa, R.C, Howard S.L, Locke R.J, Miziolek A.W, Laser – Based Flame Species Profile Measurements: A Comparison With Flame Model Predictions, Combustion and Flame, *The Journal of Combustion Institute*, Elsevier, America, 92, 1&2: 85-105 (1993).

3 Gore J.P and Zhan N.J., Nox Emission and Major Species Concentrations in partially Premixed Laminar Methane/Air Co-flow Flames, Combustion and Flame, *The Journal of Combustion Institute*, Elsevier, America, 105, 3: 414-427 (1996).

4 Jeff – Share, No Doubt About it, Natural Gas is No.1, Pipeline & Gas Journal, 231, 6: 2004, p. 40.

5 Joao Cassiano, Manuel Heitor, Antonio M and Tito Silva, Experiments in Large Scale Combustion Systems and the Characterization of the Burning Equipments. *Journal of Energy and Efficiency*, Elsevier Science Publishers Ltd. 001: 893-902 (1993).

6 Keller J.O, Bramlette T.T, Barr P.K and Alvarez J.R, NOx, and CO Emissions from a Pulse Combustor Operating in a Lean Premixed Mode. Combustion and Flame, *The Journal of the Combustion, Institute*, Elsevier, 99, ¾: 460-466 (1994).

7 Kennedy Ian M, Clement Yam, Darrell C. Rapp, and Robert J.Santoro, Modeling and Measurements of Soot and Species in a Laminar Diffusion Flame. Combustion and Flame, *The Journal of the Combustion Institute*, Elsevier, America, 107, 4: 368-382 (1996).

8 Lahaye, J. and Prado, G., *Soot in Combustion Systems and its Toxic Properties*, Plenum Press, New York, 1983.

9 Miller, J.A, Volponi, J.V and Pauwels Jean. F, The effect of Allene Addition on the structure of a Rich $C_2H_2/O_2/Ar$ Flame. Combustion and Flame, *The Journal of the Combustion Institute*, Elsevier, America, 105: 451-461 (1996).

10 Musick, M, Van Tiggelen, P.J and Vandooren, J., Experimental Study of the Structure of several Fuel-Rich Premixed Flames of Methane, Oxygen and Argon. Combustion and Flame, *The Journal of the Combustion Institute*, Elsevier, America, 105: 433-450 (1996).

11 Ofodu, J.C and Hart, H.I, Temperature and Soot Formation in a Natural Gas Diffusion Flame, Journal of Scientific & Industrial Research, India, 66: 570-576 (2007).

12 Ritter, H and Benke, G, Natural Gas for Domestic Appliances in Austria, Future Perspectives and the Potential of Energy Efficient Technologies. SAVE conference: Energy Efficiency in House hold Appliances and Lighting, Austria, 2000.

13 Tao, Y, Hodgins, K.B, and Hill, P.G, NOx Emissions From a Diesel Engine Fueled With Natural Gas, Transactions of ASME, Journal of Energy Resources Technology, 117: 290-296 (1985).

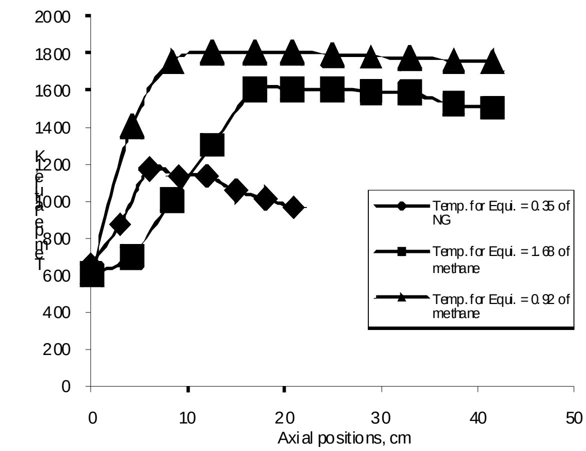

Figure 2: Scaled-up Version of Musick et al (1996) and that obtained from the in-house natural gas test rig

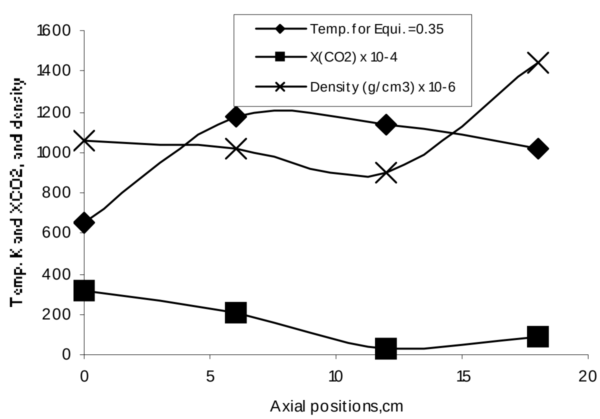

Figure 3: Axial Profile of Temperature, XCO2, and Gas density of of natural gas flame for Equivalence ratio of 0.35

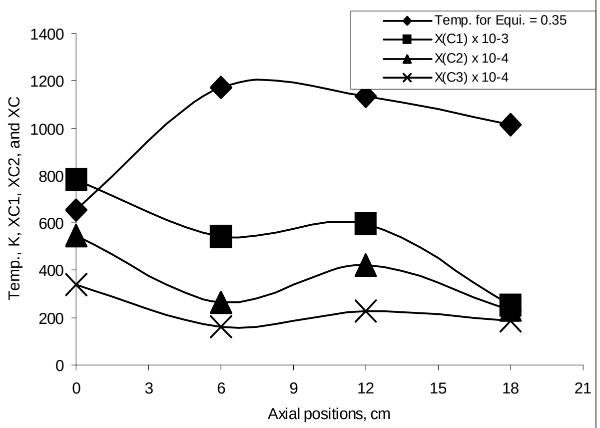

Figure 4.: Axial Profile of Temperature and XC1, XC2, and XC3
for equivalence ratio of 0.35

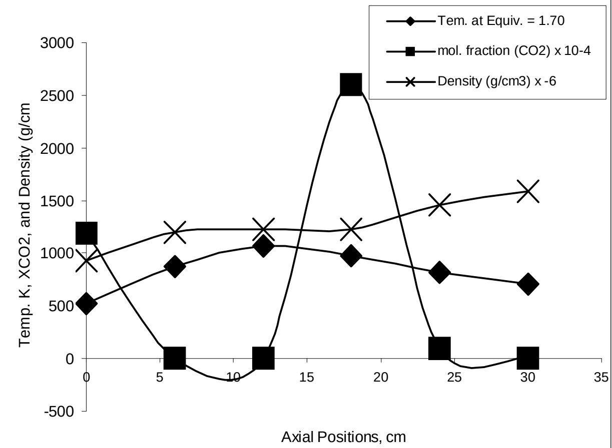

Figure 7: Axial profile of Temperature, XCO2, and Density (g/cm3) for Equivalence ratio 1.70

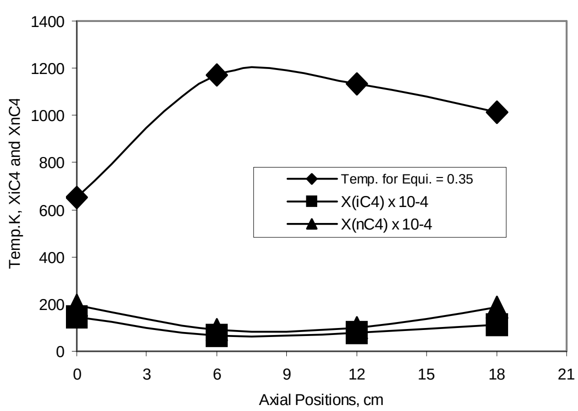

Figure 5: Axial Profiles of Temperature and XiC4 and XnC4
for Equivalence ratio 0.35

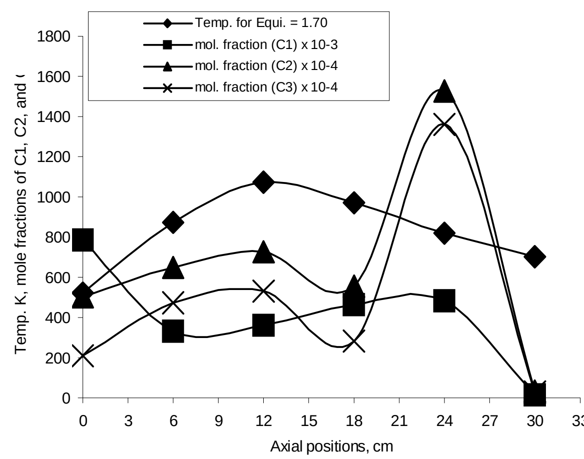

Figure 8: Axial profile of Temperature, XC1, XC2, XC3 for Equivalence ratio 1.70

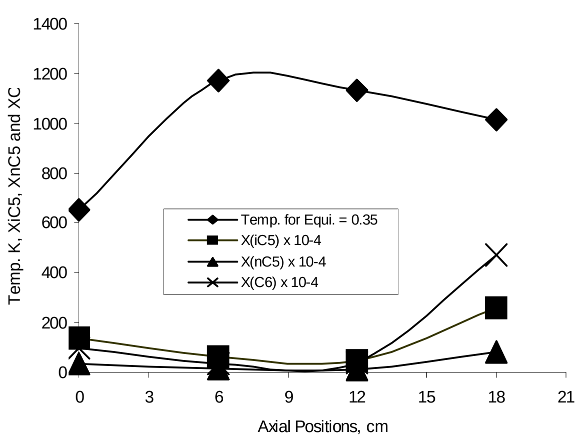

Figure 6: Axial Profiles of Temperature, and XiC5, XnC5 and XC6 for Equivalnce ratio 0.35

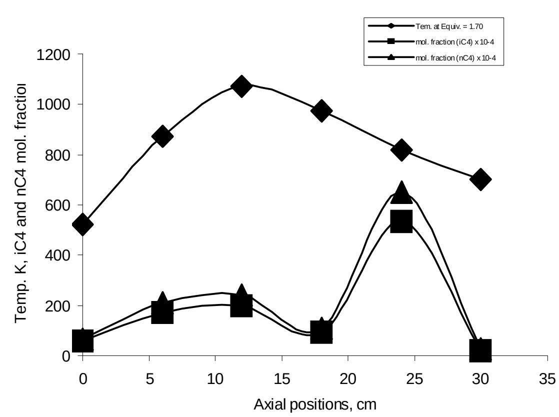

Figure 9: Axial profiles of Temperature, iC4 and nC4 mol. Fractions for Equivalence ratio 1.70

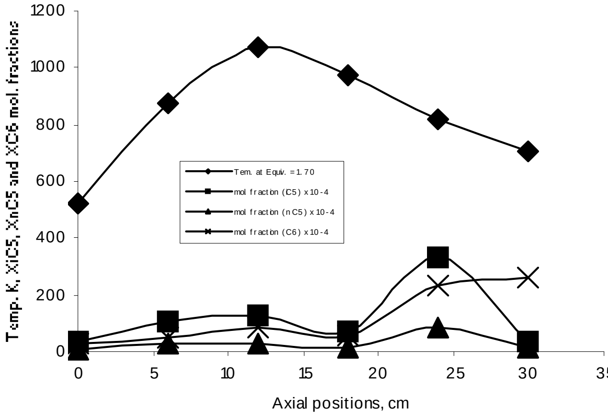

Figure 10: Axial profiles of Temperature, XiC5, XnC5, and XC6 for Equivalence ratio 170

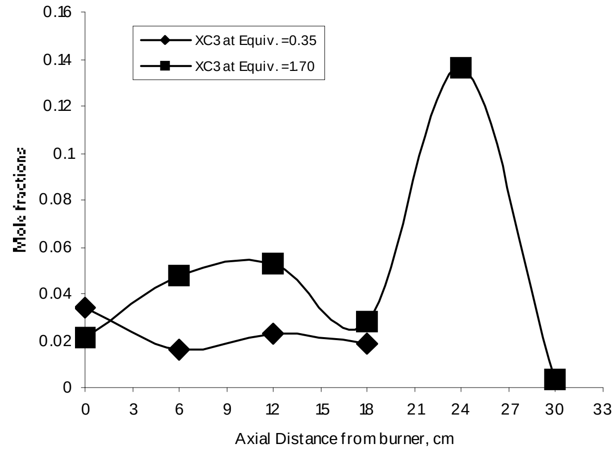

Figure 13: Axial Mole fraction profiles of C3 at Equivalence ratios of 0.35 and 170

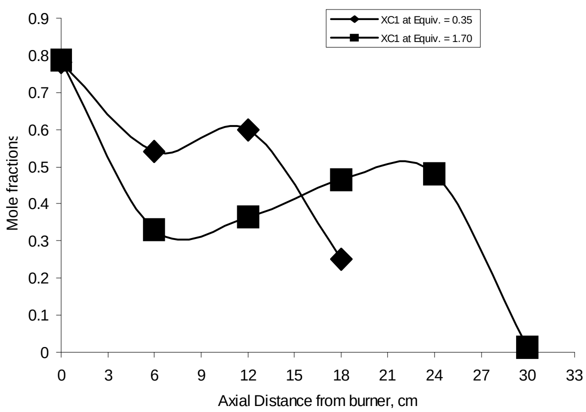

Figure 11: Axial Mole fractions profiles of C1 at Equivalence ratios of 0.35 and 1.70

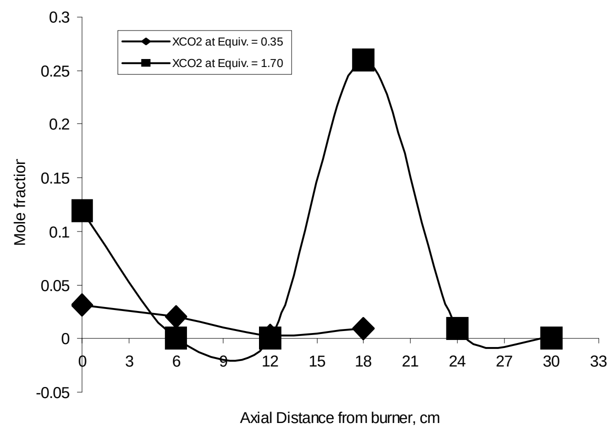

Figure 14: Axial Mole fraction profiles of CO2 for Equivalence ratios of 0.35 and 1.70

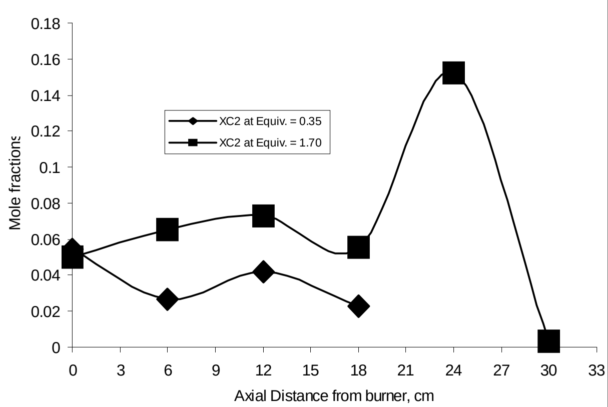

Figure 12: Axial Mole fractions profiles of C2 at Equivalence ratios of 0.35 and 1.70

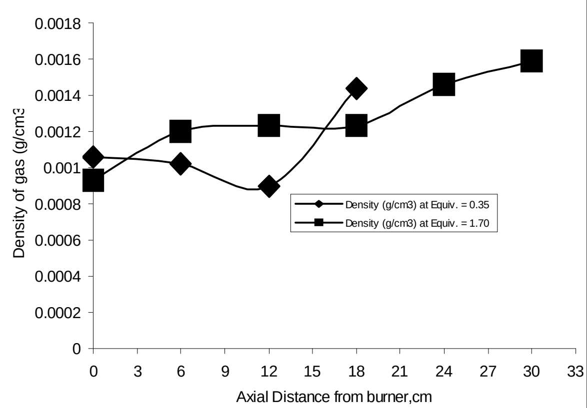

Figure 15: Axial profiles of Density of gas for Equivalence ratios of 0.35 and 1.70