NUMERICAL STUDY OF LAMINAR PREMIXED METHANE/AIR FLAMES WITH CARBON DIOXIDE DILUTION

Miguel Angel Mendieta Patiño, mmendieta@fem.unicamp.br[1]
Dario Alviso, dalviso@ing.una.py[2,3]
Rogério Gonçalves dos Santos, roger7@fem.unicamp.br [1]

[1]Faculdade de Engenharía Mecánica da Universidade Estadual de Campinas - FEM/UNICAMP, Rua Mendeleyev, 200 - CEP 13083-860, Cidade Universitária "Zeferino Vaz" Barão Geraldo, Campinas - SP
[2]Laboratorio de Mecánica y Energía, Facultad de Ingeniería, Universidad Nacional de Asunción, Campus Universitario, San Lorenzo, 2160 Paraguay
[3]Laboratorio de Fluidodinámica, Facultad de Ingeniería, Universidad de Buenos Aires, Paseo Colón 850 CABA Argentina

Abstract: *In this work carbon dioxide dilution on the laminar premixed methane/air flames was studied by means of kinetic modelling. This study contributes to the understandings of the effect of CO_2 concentrations on the combustion characteristics of biogas. The simulations were performed using the REGATH package with GRI 3.0 chemical scheme. Dilution from 0% to 50% CO_2 (volume) were computed at atmospheric pressure of 0.1 MPa an inlet temperature of 298 K with equivalence ratios from 0.7 to 1.3. Laminar flame speeds were estimated using a one-dimensional premixed flame model. The laminar flame speed and flame temperature of CH_4/air decreased with increasing CO_2 dilution. The increase in CO_2 concentrations reduced the reactants' concentrations, decreasing the net reaction rate and thus the flame speed.*

Keywords: *Biogas, Kinetic Modeling, Numerical Simulation, Premixed Flame, Dilution, CO_2*

1. INTRODUCTION

Biogas typically refers to a gas produced by the breakdown of organic matter in the absence of oxygen. It is a renewable energy source, like solar and wind energy. Furthermore, biogas can be produced from regionally available raw materials and recycled waste and is environmentally friendly and CO_2 neutral. Biogas is produced by the anaerobic digestion or fermentation of biodegradable materials such as manure, sewage, municipal waste, green waste, plant material, and crops (Corral *et al.,* 2007). Biogas comprises primarily methane CH_4 and carbon dioxide CO_2 and may have small amounts of hydrogen sulphide H_2S, moisture and siloxanes. The gases methane, hydrogen, and carbon monoxide CO can be combusted or oxidized with oxygen (Babel *et al.,* 2009).

A combustible gas/air mixture can be burned over a wide range of concentrations when either subjected to elevated temperatures or exposed to a catalytic surface at ordinary temperatures. However, homogeneous combustible gas/air mixtures are flammable, that is, they can propagate flame freely within a limited range of compositions (Zabetacis *et al.,* 1965). This defines the flammability limits of a combustible gas mixture, which are the lean or lower flammability limit (LFL) and the rich or upper flammability limit (UFL). Laminar flame speed, which is defined as the propagation speed of an adiabatic planar flame relative to the unburned gas, is an important parameter in combustion; this is because it not only affects the tendency of a flame to flashback and blow off, but also plays a role in the stability of the flame in the combustor (Chan *et al.,* 2015). Laminar premixed flames, frequently in conjunction with diffusion flames, have application in many residential, commercial, and industrial devices and processes. Examples include gas ranges and ovens, heating appliances, and Bunsen burners. Laminar premixed natural-gas flames also are frequently employed in the manufacturing of glass products (Turns *et al.,* 1996).

For any diluent, the effect of its addition could be either dilution, transport and thermal diffusions, chemical, or their combinations, as below (Zahedi *et al.,* 2014):

- Dilution effect - In the presence of diluent, the concentrations of the fuel and oxidant are reduced, leading to a lower net reaction rate and thus the lower flame speed;

- Transport and thermal effects - When a diluent is added, the mass/thermal diffusivities and the specific heat capacity of the mixture change, affecting the burning velocity;

- Kinetic effect – The diluent involves chemically, altering the reaction kinetics and consequently the burning velocity.

Di Benedetto *et al.,* 2009 studied the role of CO_2 on the flammability of $CH_4/O_2/N_2/CO_2$ mixtures and has found that the main effect of CO_2 was not on the kinetics or diffusive transport fluxes, but largely thermal, i.e. the presence of CO_2 increased the specific heat of the mixture, lowering the flame temperature and combustion rate.

Chan *et al.,* 2015 have recently published a work on to study the effect of CO_2 dilution (0-15% vol.) on the laminar flame speed of premixed methane/air mixtures, both experimentally and numerically. Persis *et al.,* 2013 studied the effects of enrichemnt and CO_2 dilution on laminar methane flames (experimentally and numerically). Results for laminar flame velocity and temperature were presented for equivalence ratio from 0.7 to 1.1 and a large range of oxygen enrichment and CO_2 dilution.

In this work, the kinetic modelling for methane oxidation in a 1D freely propagating gaseous premixed flame was performed using the REGATH package developed at EM2C laboratory (Darabiha *et al.,* 1993, Candel *et al.,* 2011, Franzelli *et al.,* 2013 and Alviso *et al.,* 2015), that takes into account the detailed kinetic and transport phenomena (heat and mass transfer) through a numerical predictive 1D code.

The aim of this work was to extend the study of Chan *et al.,* 2015 on the effect of CO_2 dilution (0-50% vol.) on the combustion characteristic of methane/air mixtures. The results would have important implications in the optimization combustor systems used for synthetic gas, particularly biogas, which is yielded from the anaerobic digestion of bio-wastes and comprises mainly methane and carbon dioxide. In a next step, the objective is to develop a new model and compare the flame speeds estimations to the experimental results.

2. LAMINAR FLAME VELOCITY

The laminar burning velocity is a physicochemical property dependent on the temperature, pressure, and mixture composition (fuel type, equivalence ratio, and amount of diluents). As a result, the laminar burning velocity provides invaluable information on the combustion properties and the underlying oxidation chemistry of the given fuel. It is also an important parameter in the design of engines, burners and other equipment where combustion is involved. Several experimental setups can be used to measure the laminar burning velocity of fuels: flat flame burners, combustion chambers, counterflow burners, etc. (Ranzi *et al.,* 1998).

It is found that a detailed knowledge of laminar premixed flames will provide insights into such properties as heat release rate, flammability limits, propagation rates, quenching and emissions characteristics. It is also common to use measured burning velocities to validate chemical kinetic schemes. Although the majority of fuel is probably burnt in turbulent combustion, data on laminar burning velocities are still needed as input to many turbulent combustion models. Also, in internal combustion engines the initial combustion is laminar, so again there is a need for the laminar burning velocity (Takashi *et al.,* 2005).

In this study, laminar flame velocities as a function of the equivalence ratio have been estimated in the case of carbon dioxide dilution on the laminar premixed methane/air flames using the 1D-PREMIXED code within the REGATH package.

3. FREELY PROPAGATING PREMIXED FLAMES GOVERNING EQUATIONS

We consider a steady, isobaric, quasi-one-dimensional flame propagation configuration. A methane/air mixture diluted in CO_2 is injected from one side, and a premixed flame propagates freely towards fresh gases with the corresponding laminar flame velocity. In order to get a stabilized flame (instead of a propagating flame) one must inject fresh gases with a velocity equal to the laminar flame velocity.

Gaseous phase are described by the following balance equations (mass, energy and species, respectively). It must be noted that there is one species conservation equation for each species present in the numerical model (N_{sp}).

$$\frac{\partial \rho_g v_g}{\partial z} = 0, \tag{1}$$

$$\rho_g v_g c_{p_g} \frac{\partial T_g}{\partial z} = \frac{\partial}{\partial z}\left(\lambda_g \frac{\partial T_g}{\partial z}\right) - \sum_{k=1}^{K} h_k W_k \Omega_k - \left(\sum_{k=1}^{K} \rho_g Y_k V_{kz} c_{p_{gk}}\right)\frac{\partial T_g}{\partial z}, \tag{2}$$

$$\rho_g v_g \frac{\partial Y_k}{\partial z} = -\frac{\partial}{\partial z}\left(\rho_g Y_k V_{kz}\right) + W_k \Omega_k, \qquad k = 1, ..., N_{sp} \tag{3}$$

The above system of equations is completed by the ideal gas equation.

The solutions have the form: gas density $\rho_g = \rho_g(z)$, gas axial velocity $v_g = v_g(z)$, gas temperature $T_g = T_g(z)$ and species mass fractions $Y_k = Y_k(z)$, $k = 1, ..., N_{sp}$ (N_{sp} is the number of species).

In these equations $c_{p_{gk}}$ and c_{p_g} are the heat capacity at local constant pressure of species k and of the mixture respectively. h_k, W_k, Ω_k are the specific enthalpy, the molar weight and the molar chemical production rate of the kth species respectively, and V_{kz} is the diffusion velocity of the kth species in the axial direction.

4. NUMERICAL SIMULATION

In this work, the dilution was expressed as a fraction of the fuel mixture. For instance, 10 % dilution with CO_2 means that the fuel is a gas mixture consisting of 90 % volume methane and 10 % volume CO_2. The stoichiometric compositions of the methane/air mixture with different levels of CO_2 dilution are summarized in Table 1.

4.1 Numerical conditions

REGATH package was used to simulate the one-dimensional model freely propagating flame premixed, developed at EM2C laboratory (Darabiha *et al.,* 1993, Candel *et al.,* 2011, Franzelli *et al.,* 2013 and Alviso *et al.,* 2015), that takes into account the detailed kinetic and transport phenomena (heat and mass transfer) through a numerical predictive one

Table 1: **The stoichiometric compositions of the methane/air mixture with different levels of CO_2 dilution**

CH_4(%vol)	CO_2 dilution(%vol)	CH_4	CO_2	O_2	N_2
100	0	0.095	0.000	0.190	0.715
90	10	0.094	0.010	0.188	0.707
80	20	0.093	0.023	0.186	0.698
70	30	0.091	0.039	0.183	0.687
60	40	0.089	0.060	0.179	0.672
50	50	0.087	0.087	0.174	0.653

dimensional code. Combustion was assumed to start at atmospheric pressure of 0.1 MPa and inlet temperature of 298 K. Equivalence ratios was varied from 0.7 to 1.3. Combustion of the methane-carbon dioxide/air mixture was simulated using the GRI 3.0 chemical scheme (Smith *et al.,* 1998), which consists of 53 species and 325 reactions. The proportions of mixture of methane and CO_2 in volume chosen were: 90% methane and 10 % of CO_2, 80% methane and 20 % of CO_2, 70% methane and 30 % of CO_2, 60% methane and 40 % of CO_2, 50% methane and 50 % of CO_2. Flame speed and maximum flame temperature were compared.

5. RESULTS AND DISCUSSION

It was simulated a 2000 mm tube. Scale ranged from -1000 mm to 1000 mm. Combustion starts at 0 mm.

Figure 1 presents a typical structure of a premixed methane flame. Mass fraction profiles of the methane/air flame were simulated using GRI 3.0 chemical scheme with equivalence ratio 1.0 and 0 % CO_2 dilution are presented. It can be seen that CH_4 and O_2 (reactants) mass fractions remain constant until a little more than 0 mm, and then combustion occurs and the reactants concentrations decrease drastically. Consequently, the products of combustion (mainly CO_2 and H_2O, and in a second place CO) are produced and starts to increase. Finally, mass fractions of these species reach a plateau, meaning that chemical equilibrium has been reached.
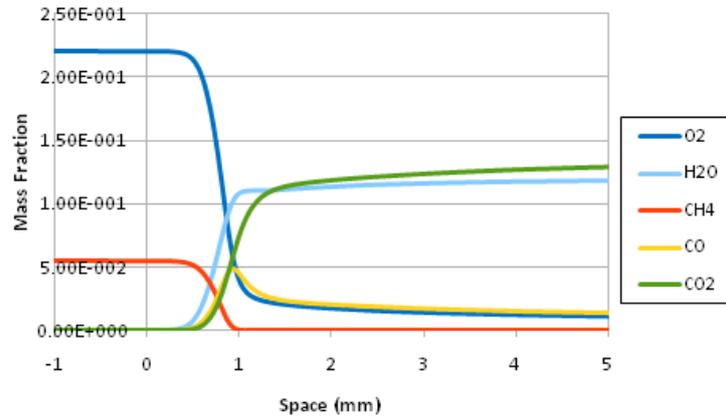

Figure 1: Mass fraction profile of reactants and products of methane/air mixture at 0.1 MPa, 298 K, equivalence ratio 1.0 and 0 % CO_2 dilution

In "Figure 2" and "Figure 3" (20% and 40% CO_2 dilution respectivaly) are observed than, besides the decrease of the concentrations of the reactants, combustion starts getting closer to 0 mm as it CO_2 dilution increases. The combustion products H_2O, and CO are both produced in small quentity as dilution of CO_2 increases.

Figure 4 shows the variation on the laminar flame speed of methane/air mixture due to the effect of CO_2 dilution, for equivalence ratios from 0.7 to 1.3. It is shown that the increase in CO_2 concentration decreased the flame speed of the methane/air mixture. Such result is consistent with the findings published in Chan *et al.,* 2015 (0-15 % vol of CO_2) and Persis *et al.,* 2013 (10-60 % of CO_2). In this study, we extend the dilution up to 50 % vol of CO_2, and it can be seen that flame velocity, after 20 % vol of CO_2 decreases drastically. From 30 % vol of CO_2, flame velocity is very far away from the results without dilution, suggesting that CO_2 dilution percentage should be limited to lower amounts. In addition, for equivalence ratios higher than 1(rich flames) the difference between diluted (from 30 % vol of CO_2) and non-diluted flames velocities is higher.

Figure 5 presents a comparison between flame temperature profiles of the methane/air mixture with equivalence ratio 1.0 and for different dilutions (0-50 % vol of CO_2). It can be seen that, near thermal equilibrium (after 4 mm) flame temperature decreases when increasing CO_2 dilution.
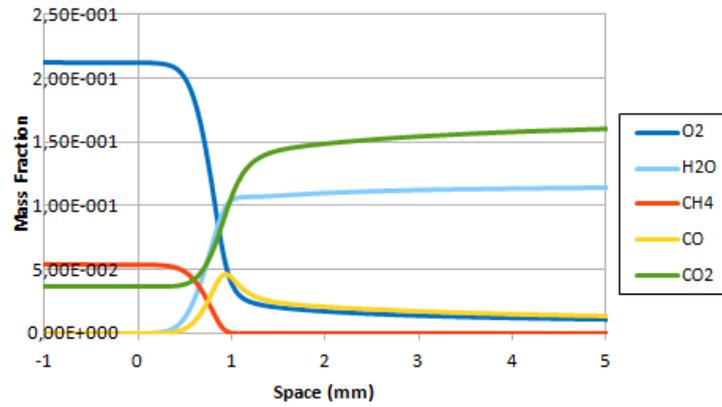
3

Figure 2: Mass fraction profile of reactants and products of methane/air mixture at 0.1 MPa, 298 K, equivalence ratio 1.0 and 20 % CO_2 dilution
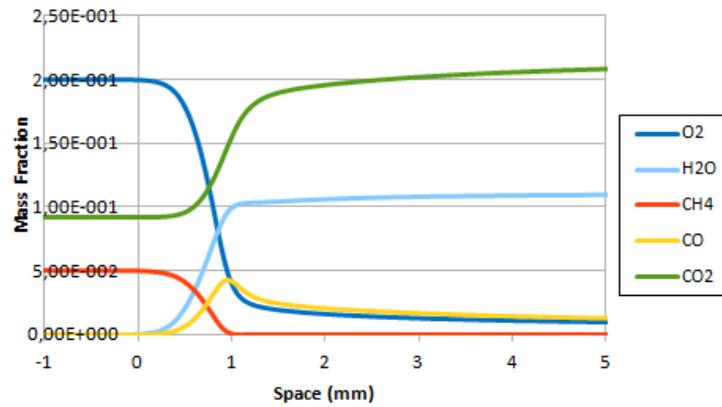

Figure 3: Mass fraction profile of reactants and products of methane/air mixture at 0.1 MPa, 298 K, equivalence ratio 1.0 and 40 % CO_2 dilution
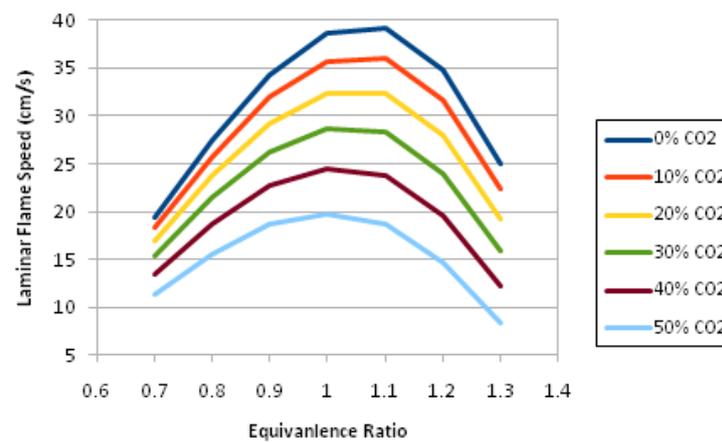

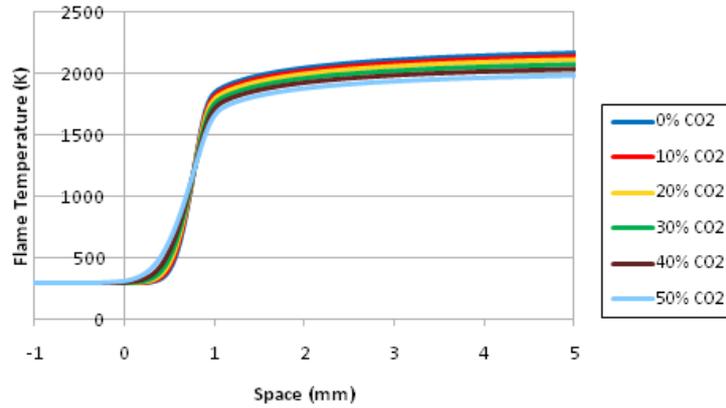Figure 4: Effect of CO_2 dilution on the flame speed profile of methane/air mixture at 0.1 MPa and 298 K.

Figure 5: Effect of CO_2 dilution on the flame temperature profile of methane/air mixture at 0.1 MPa, 298 K and equivalence ratio 1.0.

Finally, "Figure 6" extends the results of "Figure 5". In this case, maximum flame temperature of methane/air mixture for equivalence ratios from 0.7 to 1.3 and for different CO_2 dilutions (0-50 % vol of CO_2) are presented. It is shown, as expected, that the increase in CO_2 concentration decreased the maximum flame temperature of the methane/air mixture. In addition, as for the flame velocity, from 30 % vol of CO_2, maximum flame temperature is very far away from the results without dilution. And also for rich flames (equivalence ratios higher than 1) the difference between diluted (from 30 % vol of CO_2) and non-diluted maximum flame temperature is increased. The reduction in the laminar flame velocities and maximum flame temperature due the CO_2 dilution is mainly due to thermal effects (Xianzhong *et al.,* 2016).
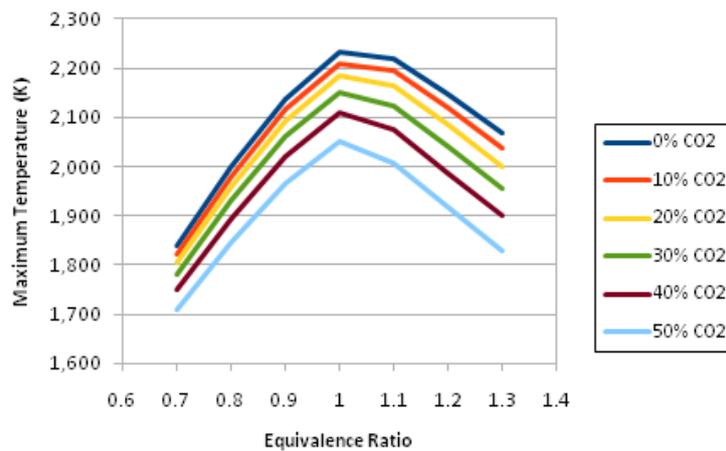

Figure 6: Effect of CO_2 dilution on the flame maximum temperature profile of methane/air mixture at 0.1 MPa and 298 K.

6. CONCLUSIONS

This paper has presented numerical studies of methane/air diluted in CO_2 combustion in a 1D freely propagating flames.

A typical flame structure of a premixed methane flame was presented and analysed. Reactants (CH_4 and O_2) and products of combustion (mainly CO_2 and H_2O, and in a second place CO) mass fractions are presented and analysed.

The influence of CO_2 dilution on the laminar flame speed of methane/air mixture was also presented, for equivalence ratios from 0.7 to 1.3 and for 0-50 % vol dilution of CO_2. It was shown that, from 30 % vol of CO_2, flame velocity is very far away from the results without dilution, suggesting that CO_2 dilution percentage should be limited to lower amounts. In addition, for equivalence ratios higher than 1 (rich flames) the difference between diluted (from 30 % vol of CO_2) and non-diluted flames velocities was increased.

Finally, maximum flame temperature of methane/air mixture for equivalence ratios from 0.7 to 1.3 and for different CO_2 dilutions (0-50 % vol of CO_2) was also presented. It was shown that from 30 % vol of CO_2, maximum flame temperature is very far away from the results without dilution.

7. ACKNOWLEDGEMENTS

8. REFERENCES

Alviso, D., Rolon, J.C., Scouflaire, P., Darabiha, N., 2015. Experimental and numerical studies of biodiesel combustion mechanisms using a laminar counterflow spray premixed flame. Fuel 153 (2015) 154-165

Babel, S., Sae-Tang, J., Pecharaply, A., 2009. Anaerobic co-digestion of sewage and brewery sludge for biogas production and land application. International Journal of Environmental Science and Technology, 6 (1): 131-140.

Candel, S., Schmitt, T., Darabiha, N., 2011. Progress in transcritical combustion: experimentation, modeling and simulation. In: 23rd ICDERS, Irvine

Chan, Y. L., Zhu, M. M., Zhang, Z. Z., Liu, P. F., and Zhang, D. K., 2015. The Effect of CO2 Dilution on the Laminar Burning Velocity of Premixed Methane/Air Flames. Energy Procedia, Vol. 75, pp. 3048–3053.

Corral, M., Argelia, M., 2007. Biogas production via anaerobic digestion of high solids livestock manures, PhD thesis, New Mexico State University, 593-599.

Darabiha, N., Lacas, F., Rolon, J., and Candel, S., 1993. Laminar counterflow spray diffusion flames: A comparison between experimental results and complex chemistry calculations. Combustion and Flame, 95 (3), pp. 261-275.

Di Benedetto, A., Di Sarli, V., Salzano, E., Cammarota, F., and Russo, G., 2009. Explosion behavior of $CH_4/O_2/N_2/CO_2$ and $H_2/O_2/N_2/CO_2$ mixtures. Int J Hydrogen Energy;34: (16) 6970-8.

Franzelli, B., Fiorina, B., Darabiha, N., 2013. A tabulated chemistry method for spray combustion, Proceedings of the Combustion Institute, Vol. 34, Issue 1, pp. 1659-1666

Persis, S., Foucher, F., Pillier, L., Osorio, V., Gokalp, I., 2013. Effects of O_2 enrichment and CO_2 dilution on laminar methane flames. Vol. 55, pp. 1055-1066

Smith GP, Golden DM, Frenklach M, Moriarty NW, Eiteneer B, Goldenberg M., et al."GRI MECH 3.0"; 1998.

Ranzi E., Frassoldati A., Grana R., Cuoci A., Faravelli T., Kelley A. P., Hierarchical and comparative kinetic modeling of laminar flame speeds of hydrocarbon and oxygenated fuels, Prog. Energy Combust. Sci., 38 (2012) 468-501. Flame, 115 (1998) 126-144.

Takashi, H. and Kimitoshi, T., 2005. Laminar Flame Speeds of Ethanol, N-Heptane, Isooctane air Mixture. Department of Mechanical Engineering, Oita University, Japan

Turns, S.R. et al., 1996. An introduction to combustion, Vol. 287. McGraw-hill New York.

Xianzhong, H., Qingbo, Y., Junxiang, L., 2016. Chemical effect of CO2 on the laminar flame speeds of oxy-methane mixtures in the condition of various equivalence ratios and oxygen concentrations. Vol. 41, pp. 15068-15077.

Zabetacis, M. G., 1965. Flammability Characteristics of Combustible Gases and Vapors. U.S. Department of the Interior, Bureau of Mines.

Zahedi, P., and Yousefi, K., 2014. Effects of Pressure and Carbon Dioxide, Hydrogen and Nitrogen Concentration on Laminar Burning Velocities and NO Formation of Methane-Air Mixtures. J Mech Sci Technol;28: (1) 377-86.

9. RESPONSIBILITY NOTICE

The authors are the only responsible for the printed material included in this paper.