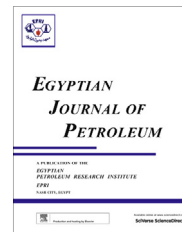




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FULL LENGTH ARTICLE

Fuel rich and fuel lean catalytic combustion of the stabilized confined turbulent gaseous diffusion flames over noble metal disc burners



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Abstract Catalytic combustion of stabilized confined turbulent gaseous diffusion flames using Pt/Al₂O₃ and Pd/Al₂O₃ disc burners situated in the combustion domain under both fuel-rich and fuel-lean conditions was experimentally studied. Commercial LPG fuel having an average composition of: 23% propane, 76% butane, and 1% pentane was used. The thermal structure of these catalytic flames developed over Pt/Al₂O₃ and Pd/Al₂O₃ burners were examined via measuring the mean temperature distribution in the radial direction at different axial locations along the flames. Under-fuel-rich condition the flames operated over Pt catalytic disc attained high temperature values in order to express the progress of combustion and were found to achieve higher activity as compared to the flames developed over Pd catalytic disc. These two types of catalytic flames demonstrated an increase in the reaction rate with the downstream axial distance and hence, an increase in the flame temperatures was associated with partial oxidation towards CO due to the lack of oxygen. However, under fuel-lean conditions the catalytic flame over Pd catalyst recorded comparatively higher temperatures within the flame core in the near region of the main reaction zone than over Pt disc burner. These two catalytic flames over Pt and Pd disc burners showed complete oxidation to CO₂ since the catalytic surface is covered by more rich oxygen under the fuel-lean condition.

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1. Introduction

Catalytic combustion or heterogeneous combustion had been extensively investigated in recent years. The catalytic oxidation of hydrocarbons became the focus of much basic and applied catalysis research because of its increasing importance for burner's design of industrial furnaces and the techniques of power-generating gas turbines [1,2]. For these applications, high temperature catalytic combustion was regarded as highly efficient and clean energy system. It had been recognized that

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noble metals possessed the highest catalytic activities that initiated the catalytic oxidation of fuels at relatively lower reaction temperatures [3,4].

Many experimental investigations reviewed the effect of using noble metals in the catalytic combustion domain. In a confined turbulent flame the effect of using Pt/ γ -Al₂O₃ catalytic disc burner was examined at different axial distances over the fuel jet nozzle [5]. This investigation revealed how the highly efficient oxidative exothermic reactions in contact with the active surface of platinum burner greatly enhanced the heat evolved via catalytic ignition and hence, improved the stabilization tendency to a great extent. Also, the progress of the combustion process over the platinum sites had controlled the combustion emission products therefore minimizing the environmental pollution. Moreover, Appel et al. investigated the catalytically stabilized turbulent combustion of fuel-lean hydrogen/air pre-mixtures over platinum and found that nearly half of the fuel was converted heterogeneously and the remaining part was combusted in the post-catalyst [6].

More recently, the catalytic combustion of different hydrocarbon fuels over platinum addressed the interaction between homogenous and heterogeneous reactions. The experimental study revealed that in the presence of platinum the reactions are complex and highly dependent on the fuel used as well as other parameters such as temperature, equivalence ratio and Reynolds number of the gaseous fuel–air mixture [7]. Furthermore, the effect of catalytic combustion of gaseous turbulent diffusion flame over a series of noble metal disc burners (Pt, Pd and Pt + Pd) supported on γ -Al₂O₃ were experimentally and mathematically studied [8]. These catalytic flames behave in highly catalytic conditions and their catalytic enhancement was found to be in the order: (Pt + Pd) > Pt > Pd. The thermal distribution along these catalytic flames recorded high values due to the enhancing of fuel oxidation on the noble metal sites in the reaction zone of the flame via improving homogeneous gas/heterogeneous surface reactions in the combustion domain.

Fuel-rich catalytic combustion had been investigated in order to demonstrate the successful technology of ultra-low emissions for gas turbine which in the mean time, offered multiple advantages, [9–12]. The catalytic combustion of methane under lean and rich conditions over platinum and palladium catalysts was investigated using dilute mixtures [13]. It had been found that under lean conditions Pd was the more effective catalyst. Pt containing catalysts had been found to be more active as the reactant mixture was shifted from oxygen–rich to methane–rich. The platinum catalysts were superior to palladium in a fuel-rich gas mixture. Thus, platinum had a role as a component in the catalyst for emission control of natural gas vehicles. Also, the performance data of catalytic combustion were presented for methane oxidation over platinum group catalysts under fuel-rich and fuel-lean conditions [14]. These authors found that under fuel-lean conditions, Pd catalyst was the most active, although deactivation occurred above 650 °C. While under fuel-rich conditions, Pt catalyst was more active above 600 °C and acquired much higher activity of the reaction rates through the catalytic combustion domain.

A rich catalytic lean burn combustion system was developed for the operation of natural gas as fuel and other non-methane fuels [15]. For fuel-rich operation the reactor performance was insensitive to the fuel reactivity, because the reaction rate (heat release) upon the catalyst surface was

controlled primarily by the oxygen mass transfer to the catalyst under fuel-rich condition and not by the fuel flow or the fuel reactivity for all the fuels tested.

For methane or natural gas fuels, the catalyst activity was significantly improved by operating the catalyst under fuel-rich conditions as compared to fuel-lean conditions and therefore, allowing a wider choice of catalyst materials. Fuel-rich methane combustion over Rh-La-MnO₃ honeycomb catalysts was developed as a preliminary conversion step in advanced combustion system such as power turbine and utility burners for reducing emissions [16]. The experimental results showed that mixed Rh-La-MnO₃ catalysts were suitable for the fuel-rich applications. However, a progressive reduction of light-off temperature and a parallel improvement of the catalytic partial oxidation performance were observed by increasing Rh content in the Rh-La-MnO₃ catalysts.

The present work investigated the effect of using Pt/ γ -Al₂O₃ and Pd/ γ -Al₂O₃ catalytic disc burners situated in the combustion domain of confined turbulent stabilized gaseous diffusion flames under fuel-rich and fuel-lean conditions. The thermal structure of these catalytic flames developed over Pt and Pd catalytic disc burners was examined by measuring the mean temperature distribution in the radial direction at different axial locations along the flames. Also, the axis-symmetric distributions of CO and CO₂ along the flames were monitored under the same conditions to clarify the catalytic combustion process performance over the two current catalytic discs.

2. Experimental

The experimental setup (Fig. 1) was comprised of a vertical cylindrical combustion chamber filled with an arrangement supplying fuel and air. The combustion chamber (Fig. 2) is 150 mm in diameter, 5 mm thick and 1.0 m long. The combustor was equipped with a thermal resistant glass window. The fuel jet was discharged vertically through a nozzle of 2.5 mm diameter connected at the centre of the fuel supply line in the axial direction at the base of the combustor. Commercial LPG fuel having an average composition of: 23% propane, 76% butane, and 1% pentane was used in all experiments.

Two catalytic disc burners of Pt and Pd over γ -Al₂O₃ having a diameter of 40 mm, 4 mm thick and perforated with 25 holes had been separately used as a catalytic flame burner. The discs were placed at the base of the combustor at the specified supporting distance of 40 mm over the fuel jet nozzle. These discs were made of γ -Al₂O₃ with a surface area of 60 m² g^{−1}. Each Al₂O₃ disc support was made by mixing γ -Al₂O₃ powder with a suitable binder then pasted and formed. After drying at 110 °C overnight, the disc was perforated by drilling to acquire a suitable perforation (3 mm holes diameter). The disc was then calcined at 400 °C for 4 h. in a muffle furnace. The heat treatment gave the highest crushing strength while retaining the catalytic activity.

The 1st disc (Pt/ γ -Al₂O₃) was impregnated with H₂PtCl₆ solution such that the Pt content was 0.0001 wt% of the disc. The disc was again dried at 110 °C overnight and calcined at 550 °C for 4 h.

The 2nd disc (Pd/ γ -Al₂O₃) was prepared via wet impregnation of an aqueous solution of Pd (NO₃)₂ containing 10^{−4} g of Pd metal. The impregnation was adjusted to incorporate the Pd containing solution on the external surface (1 mm depth).

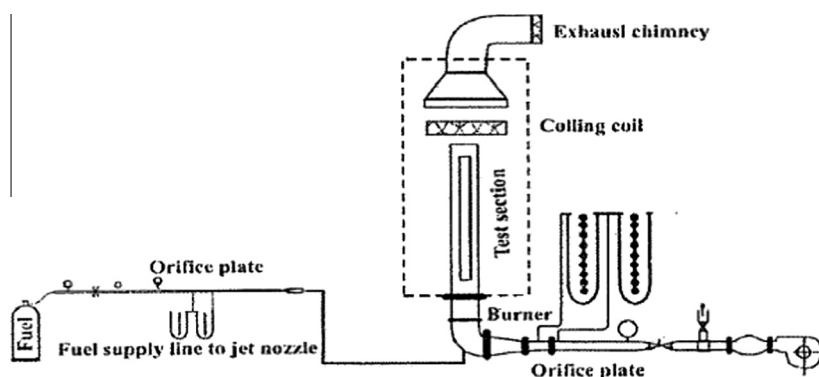


Figure 1 Experimental set up.

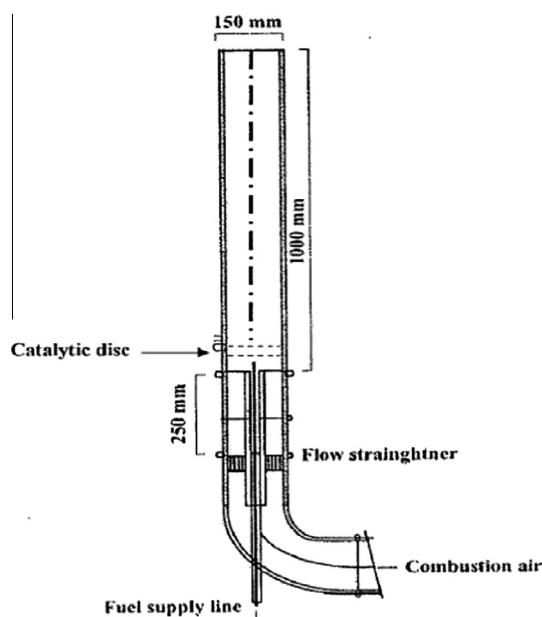


Figure 2 Combustor and burner arrangements.

Each catalytic disc was then held at the combustor position in each experiment.

Detailed local measurements of the mean temperature were examined along the stable flames in the presence of each catalytic disc burner. The temperature measurements had been obtained by using a thermocouple probe (platinum/platinum–13% rhodium, 0.1 mm diameter wire). The thermocouple output voltage was integrated over a period of 20 s using a microprocessor integrator. Also, the axial distribution of CO and CO₂ concentrations had been achieved along the flame developed over the catalytic disc burners. The concentrations were obtained through the use of stainless steel water cooled sampling probe with 1.0 mm inner diameter and 6.0 mm outer diameter. Samples of the combustion products were conditioned and measured using on-line gas analyzer (Anapol EU-200/4).

For each catalytic disc burner, the developed flame was tested under both fuel-rich and fuel-lean conditions. These test conditions were usually defined in the combustion literature by the term of overall equivalence ratio, ϕ , which was usually defined in the combustion literature as:

$$\text{Overall Conventional } \phi = \frac{\left(\frac{m\text{-fuel}}{m\text{-air}}\right)_{\text{experimental}}}{\left(\frac{m\text{-fuel}}{m\text{-air}}\right)_{\text{stoichiometry}}} = 1$$

A modification was introduced by Schmidt and coworkers [17] including the modified equivalence ratio as:

$$\text{Modified } \phi = \frac{\phi}{1 + \phi}$$

This modification had the advantage that it considered equal weight on the fuel-lean and the fuel-rich sides of the ignition condition. Unlike the overall conventional equivalence ratio ϕ , the modified equivalence ratio ϕ was bounded between 0 and 1.0, and was symmetric about the stoichiometric point (0.5) having an equal range of values on both the fuel-lean ($\phi < 0.5$) and fuel-rich ($\phi > 0.5$) sides. Two gas compositions were selected for testing, one at ($\phi = 0.25$) in the “middle” of the fuel-lean “mixture composition space”, and the other at ($\phi = 0.75$) in the “middle” of the fuel-rich mixture composition space.

3. Results and discussion

The performance data are presented in Figs. 3–10 for the catalytic combustion of stabilized gaseous turbulent diffusion flames on Pt/ γ -Al₂O₃ and Pd/ γ -Al₂O₃ catalytic disc burners under both fuel-rich and fuel-lean conditions. The thermal structure of the stabilized catalytic flames was determined via measurements of the temperature profile at different axial locations over the catalytic discs. The temperature profiles were characterized by steep gradients due to chemical combustion (oxidation) reactions and flame boundary layer where the heat transfer occurred.

Results of the mean temperature distributions of the stabilized turbulent diffusion catalytic flames developed over Pt/ γ -Al₂O₃ and Pd/ γ -Al₂O₃ catalysts in the radial direction under fuel-rich condition at the modified equivalence ratio ($\phi/1 + \phi = 0.75$) are shown in Figs. 3 and 4.

In Fig. 3, the thermal structure of the stabilized catalytic flames was developed over Pt/Al₂O₃ under fuel rich condition at $\phi/1 + \phi = 0.75$ and had been characterized by the radial distribution of the measured mean temperature at different axial distances over the disc. The results showed comparatively high temperature values within the flame core at the near upstream distance within the central flame region, at

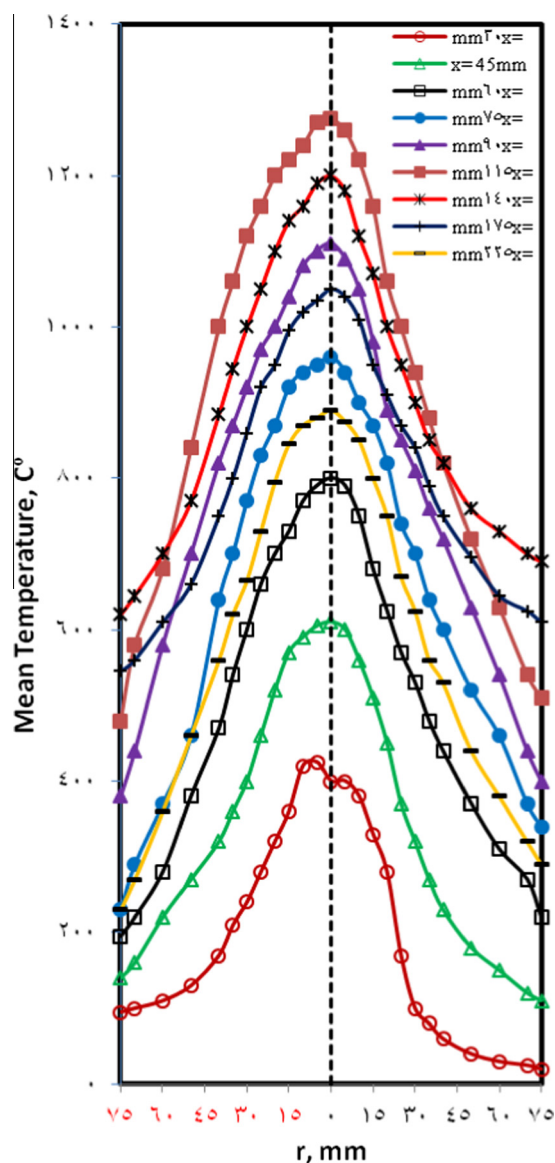


Figure 3 Radial mean temperature distribution over Pt/Al₂O₃ disc for fuel-rich condition, $\phi/1 + \phi = 0.75$ at different axial distance x .

$X = 30$ mm indicating 400 °C with respect to Pd disc (Fig. 4). The temperature profile attained maximum values at the edges of this zone then dropped radially outwards. Evidently, under fuel-rich mixture, the flame core was hot at the early upstream region in the main reaction zone of the stabilized turbulent combustion domain. A gradual increase of the axial location over the Pt catalytic disc, the radial mean temperature distribution along the flame expressed the progress of the combustion process demonstrating much higher activity. A higher reaction activity under fuel-rich condition could be assumed to provide reduced catalyst light-off temperature for catalytic flames operating over the Pt disc.

In Fig. 4, the radial mean temperature distribution of the catalytic stabilized flame over the Pd disc at different axial locations proceeded in the same trend as in the use of Pt disc but showed comparatively lower activity. At the early upstream distance over Pd disc the temperature recorded

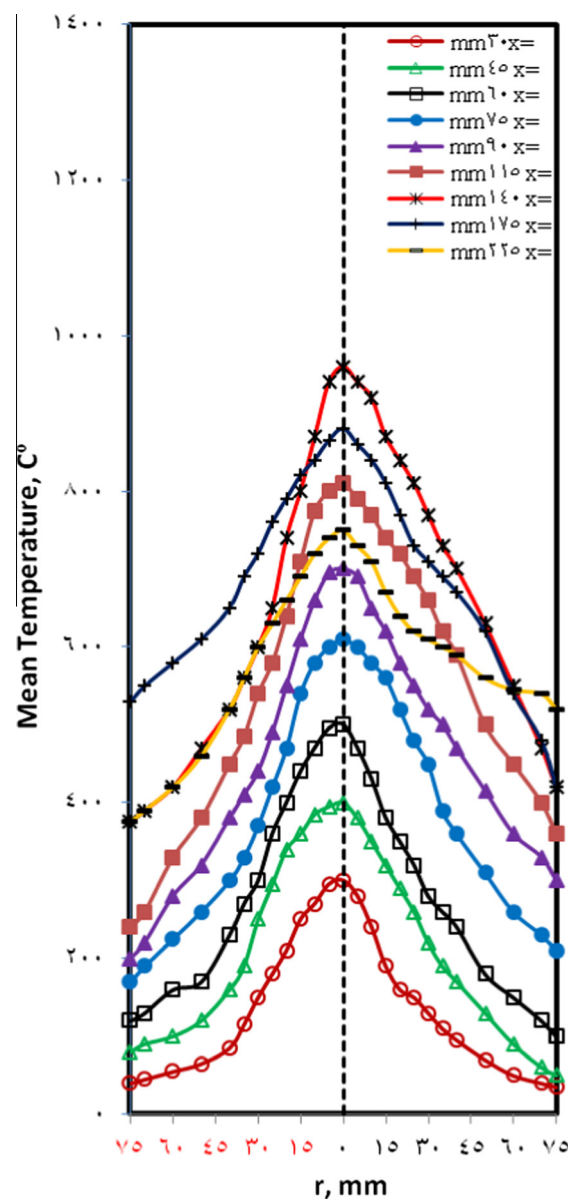


Figure 4 Radial mean temperature distribution over Pd/Al₂O₃ disc for fuel-rich condition, $\phi/1 + \phi = 0.75$ at different axial distance x .

300 °C at $X = 30$ mm within the flame core. However, the chemical reactions of the combustion process proceeded with lower active rates over the Pd disc in the combustion application under fuel-rich condition.

Fig. 5 shows the axial mean temperature distribution of the catalytic stabilized flames developed over Pt/Al₂O₃ and Pd/Al₂O₃ catalytic disc burners under fuel-rich condition, ($\phi/1 + \phi = 0.75$). The temperature distribution of these two stabilized catalytic flames exhibited similar trends; a gradual increase of temperature as a function of the axial distance, reached a maximum due to the progress of the combustion followed by a gradual decline. The maximum mean temperature of the catalytic flame developed over Pt is about 1280 °C at $X = 115$ mm whereas the maximum mean temperature of catalytic flame over Pd was about 980 °C at $X = 140$ mm. It was

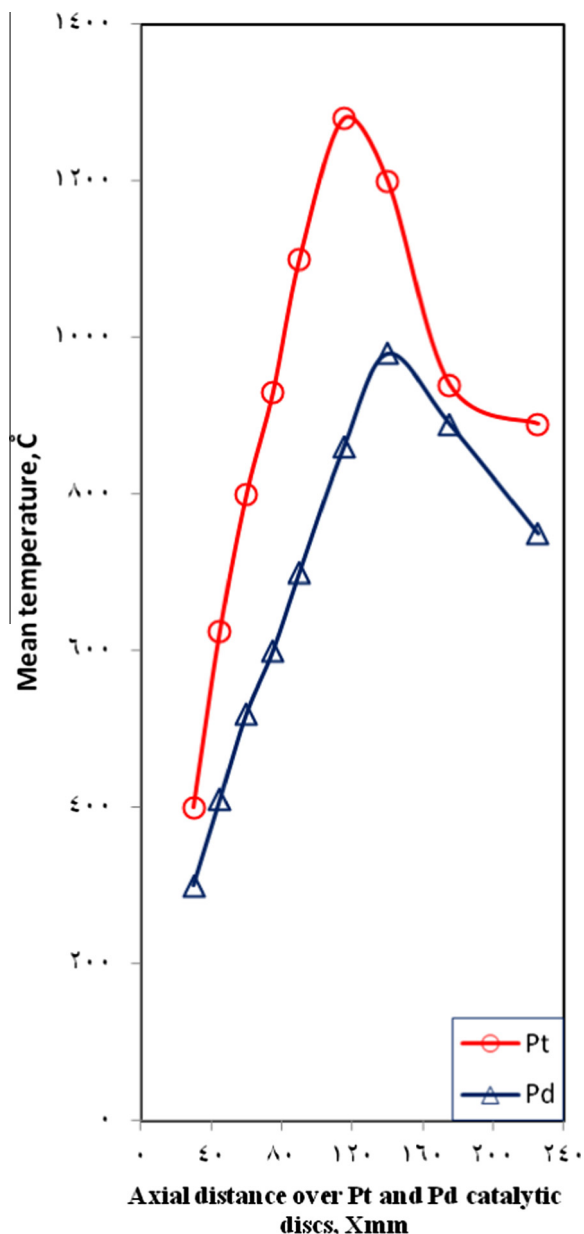


Figure 5 Axial mean temperature distribution vs. axial distance at fuel-rich condition, $\phi/1 + \phi = 0.75$.

evident that the chemical reaction proceeded with lower rates over Pd disc reaching the maxima at a further downstream distance in the catalytic combustion domain.

Fig. 6 shows the axial distribution of, CO%, at different axial locations along the stabilized flames over the two catalytic discs under fuel-rich condition ($\phi/1 + \phi = 0.75$). Pt and Pd catalysts showed the highest activity to partial oxidation to CO. The lack of oxygen over the catalysts provided CO which correspondingly increased with increasing the temperature as shown in Fig. 5. As the catalyst temperature increased the CO desorption rate increased which manifested the increase of the functionalization towards the partially oxidized product, CO.

Under the fuel-lean condition at a modified equivalence ratio ($\phi/1 + \phi = 0.25$), the radial mean temperature distribution for the two stabilized catalytic flames operated over

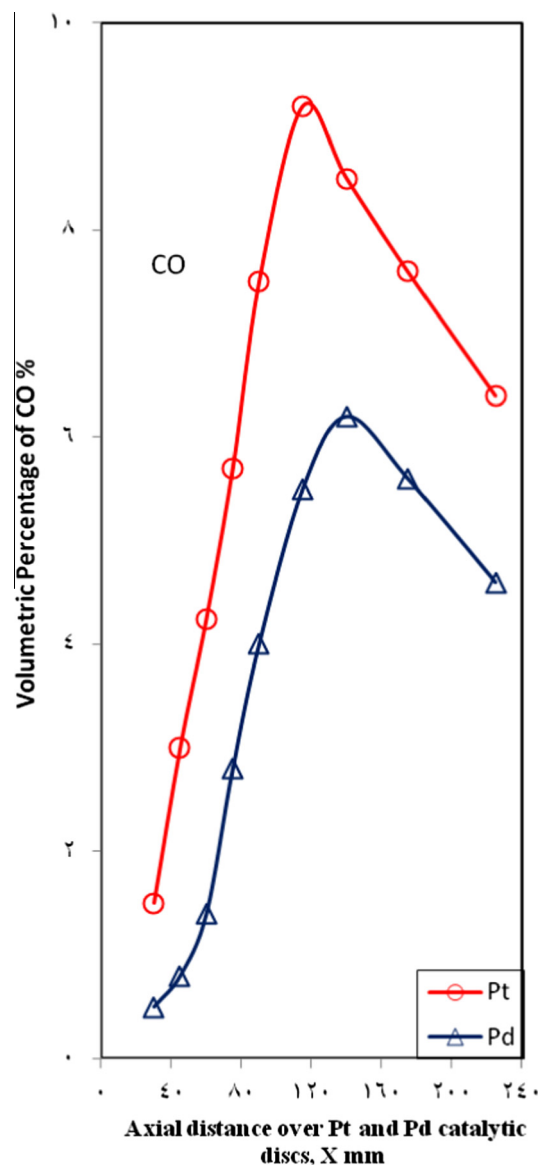


Figure 6 Axial distribution of CO% vs. axial distance at fuel-rich condition, $\phi/1 + \phi = 0.75$.

Pt/ Al_2O_3 and Pd/ Al_2O_3 disc burners at different axial locations are shown in the Figs. 7 and 8, respectively. Over the Pt disc burner, (Fig. 7), the catalytic flame was acquired at near upstream locations, $X = 30, 45, 60, 75$, the main reaction zone enclosed the flame core and was characterized by comparatively low temperature due to the presence of the unreacted mixture under the fuel-lean condition. Because of the excess oxygen, the activation of the fuel on the catalyst surface was a limiting step, while the adsorption of oxygen is fast and proceeded without significant activation barrier. The catalyst surface was therefore, covered by oxygen as adsorbent of O_2 molecules on the Pt particles. Some forms of metal oxide species could be assumed to contribute also to this reaction. Nevertheless, the temperature profiles at the downstream locations indicated a gradual increase in the main values. Because Pt had been found to be activated above 600°C [5], a gradual increase of the reaction rate was observed, i.e. the thermal energy increased.

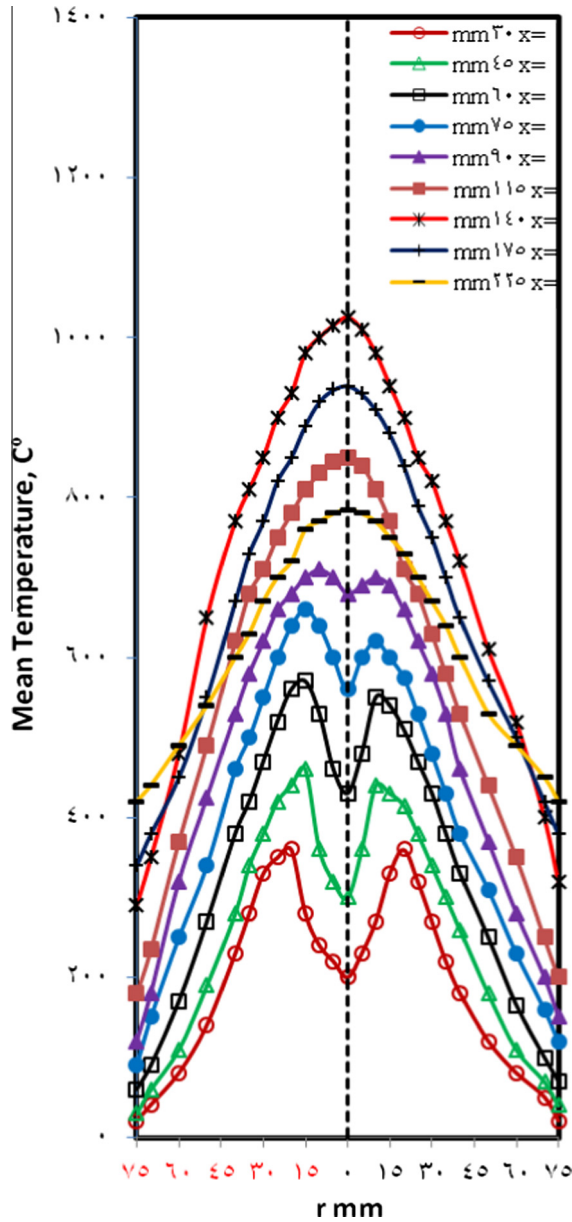


Figure 7 Radial mean temperature distribution over Pt/Al₂O₃ disc at fuel-lean condition, $\phi/1 + \phi = 0.25$ at different axial distance x .

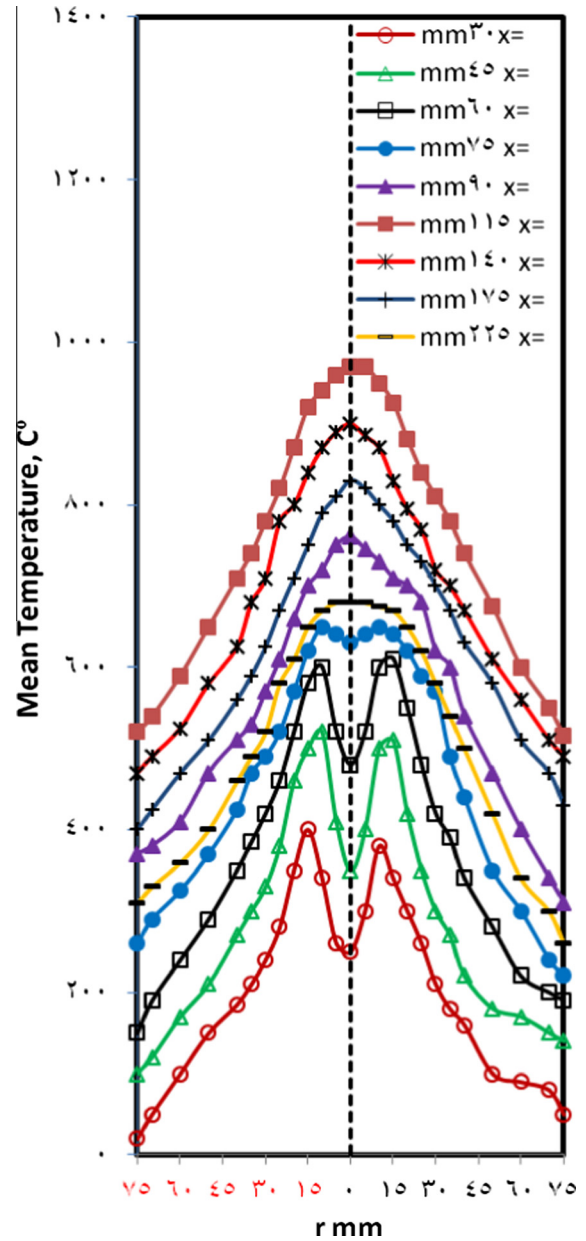


Figure 8 Radial mean temperature distribution over Pd/Al₂O₃ disc at fuel-lean condition, $\phi/1 + \phi = 0.25$ at different axial distance x .

On the other hand, Fig. 8 represents the radial mean temperature distribution of the two stabilized catalytic flames over Pd disc burner at different axial locations under fuel-lean condition ($\phi/1 + \phi = 0.25$). A closer inspection of the results revealed that the mean temperature values at early upstream distance within the flame core were comparatively higher within the main reaction zone as compared to the catalytic flame over Pt disc in the same case of fuel-lean condition. The Pd catalyst was the most active under this lean condition, although, Lyubovsky et al. [14], reported deactivation above 650 °C, i.e. the reaction rate decreased.

Fig. 9 includes a relation of the axis-symmetric mean temperature distribution of the two stabilized catalytic flames

developed over Pt and Pd disc burners at different locations under fuel-lean condition ($\phi/1 + \phi = 0.25$). At these conditions, the two flames behaved in the same manner, a gradual increase of temperature with respect to the axial distance; reached a maximum followed by a gradual decay. The mean temperature values for the flames operated over Pd catalytic disc burner at upstream distances from $X = 30$ up to 75 mm recorded higher values than the flames over Pt disc. Pd catalyst was the most active at fuel-lean condition. Lyubovsky et al. [14] confirmed this result especially within the range up to 650 °C. This is remarkably noticed within the flame core at the main reaction zone. A maximum temperature of 960 °C

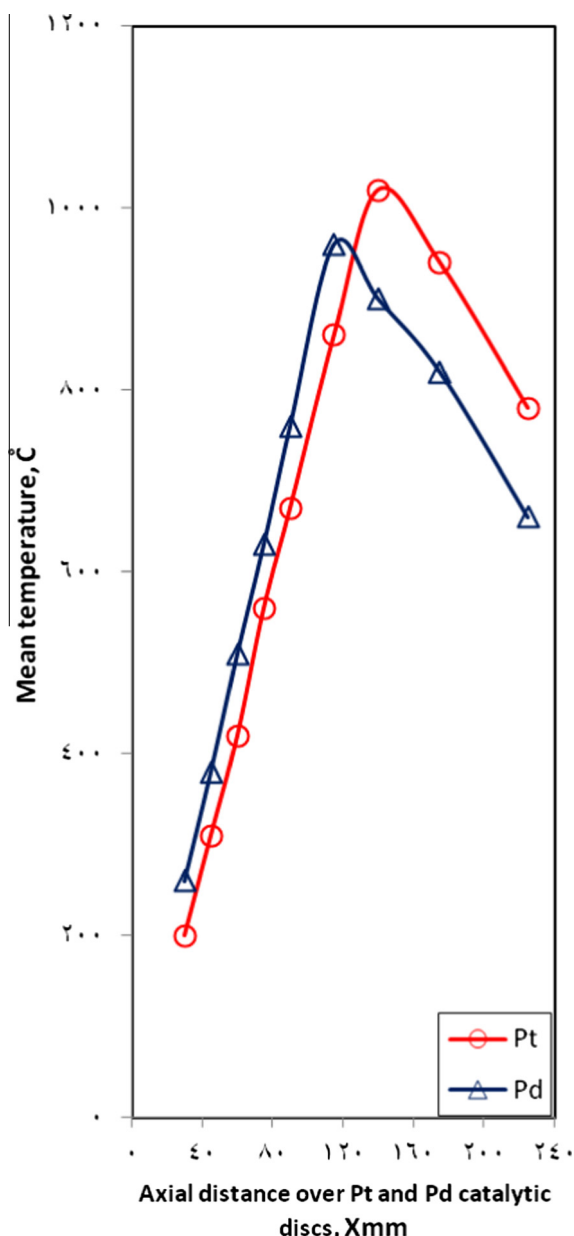


Figure 9 Axial mean temperature distribution vs. axial distance at fuel-lean condition, $\phi/1 + \phi = 0.25$.

is attained at $X = 115$ mm followed by gradual decay up to 225 mm. The recorded maximum temperature along the flame over Pt disc is 1025 °C at $X = 140$ mm. Smith et al. [15] reported that Pt is active above 600 °C in all conditions.

Fig. 10 shows the axial distribution of $\text{CO}_2\%$ along the two stabilized catalytic flames at different axial distance over Pt and Pd disc burner under fuel-lean condition ($\phi/1 + \phi = 0.25$). The axial distribution of $\text{CO}_2\%$ behaved in a similar manner as the axial thermal distribution in Fig. 9. Since the catalytic surface was covered by excess oxygen under fuel-lean conditions, the fuel dissociated with high rates and immediately was oxidized to CO_2 and H_2O .

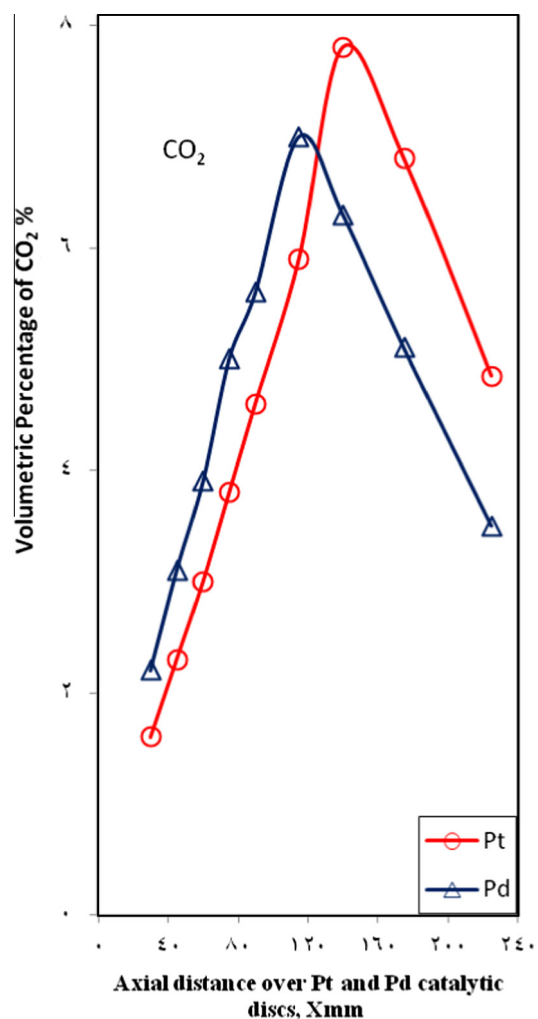


Figure 10 Axial distribution of $\text{CO}_2\%$ vs. axial distance at fuel-lean condition, $\phi/1 + \phi = 0.25$.

4. Conclusions

The following conclusions were derived from the obtained results:

- 1- The thermal structure of the stabilized turbulent gaseous diffusion flames developed over two catalytic disc burners, Pt/ $\gamma\text{-Al}_2\text{O}_3$ and Pd/ $\gamma\text{-Al}_2\text{O}_3$ indicated higher activity under fuel-rich than under fuel-lean conditions.
- 2- The catalytic flame operating over Platinum disc burner attained comparatively higher temperature values at early upstream region of the main reaction zone and reached the peak thermal values at nearer axial location as compared to the flame developed over Palladium disc burner under fuel-rich condition.
- 3- The stabilized flames over Pt and Pd catalytic discs showed high activity towards partial oxidation to CO due to the lack of oxygen over the disc, which were increased with increasing the temperature under fuel-rich conditions.

- 4- Under fuel-lean conditions, the mean temperatures at early upstream distance within the catalytic flame core operating over the Pd disc burner were higher as compared to the catalytic flames over the Pt disc.
- 5- The two catalytic flames over Pt and Pd disc burners showed the complete oxidation to CO₂ since the catalytic surface was covered by excess oxygen under fuel-lean condition.

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