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Experimental Investigation of Catalytic Surface Reaction for Different Metal Surfaces

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Abstract

The use of a catalytic surface to enhance chemical reaction rates is a well established and common practice. However, its use in combustion devices for enhancing combustion reaction is somewhat less common and more recent. Catalytic combustors, because of their inherent ability to operate at very lean air fuel mixtures, can maintain a relatively low combustion temperature and hence reduce the formation of NO_x significantly. Further the catalytic coating on the combustion chamber walls enhances the combustion process by increased rate of pre-flame reactions. This provides a basis for catalytic combustion in lean burn engine. A considerable amount of effort has been devoted in the present work to this experimental study and compares different catalyst reaction performance. A cylindrical chamber is fabricated and air-fuel mixture is passed through the chamber. A metal tube coated with the catalyst is placed inside the control volume and it is heated by an electric heater. The air-fuel mixture undergoes pre-flame combustion reaction and as a result of this, the miniature temperature increases. Various catalytic surfaces like mild steel, Nickel, Chromium and copper were tested. The activation temperatures of these catalytic surfaces were obtained from this experimental work.

Keywords: catalytic reaction, LPG, activation energy, catalytic coating

1. Introduction

Internal combustion (IC) engines are used in a variety of stationary applications ranging from power generation to inert gas production. Both spark ignition and compression ignition engines can be found. Depending on the application, stationary IC engines range in size from relatively small (~5 Hp) for agricultural irrigation purposes to thousands of horsepower for power generation. Often when used for power generation, several large engines will be used in parallel to meet the load requirements.

The operation of IC engines results in the emission of hydrocarbons (UBHC), carbon monoxide (CO), nitrogen oxides (NO_x), and particulate matter (PM). The actual concentration of these criteria pollutants varies from engine to engine, mode of operation, and is strongly related to the type of fuel used.

Various emission control technologies exist for IC engines which can offer substantial reductions in pollutants listed above. However depending on whether the engine is being run rich, lean, or stoichiometrically and the emission control technology used, the targeted emissions vary as do the levels of control.

Lean mixture operation is one of the promising method for reducing emissions and improving fuel economy in spark ignition engines. The problems associated with lean combustion are low flame velocity, combustion instability, misfire and cyclic variation of combustion . The above problems can be minimized by different techniques namely, increasing air movement, charge stratification, catalytic activation, increasing compression ratio and modifying combustion chamber. Among these methods, catalytic activation offers a simple and effective solution.

Karim and Kibrya (1986) have done detailed experimental work to compare the catalytic activation of eight different metals and found out that platinum and copper showed better performance. The catalysts in the form of wire mesh were placed in a cylindrical chamber in this study and recommended that further improvement could be achieved if the catalyst was coated inside the combustion chamber. The catalysts offered improvement in lean blowout limit. The effectiveness of various catalysts tested in the lean combustion of methane was in the order: Pt > Cu > Ag > brass > Cr > Ni > stainless steel. It was also confirmed that hydrogen was more sensitive to catalytic effects than methane.

In catalytic reaction, because chemical reactions only occur at catalyst surfaces, the location of the heat source is fixed. Hence, combustor heat transfer design is simpler than with gas-phase combustion in which reaction zone locations may change in undesirable ways. Also, generally catalytic combustion can be sustained at lower temperature than gas-phase combustion, which reduces heat loss and thermal stress problems. Furthermore, in catalytic combustors the higher surface area to volume ratio increases the flux of reactants to the catalyst surface, which could help overcome the larger heat losses and make catalytic combustion even more attractive.

Ezekoye et al. (1992) carried out experiments on catalytic surfaces using propane (C₃H₈) and methane (CH₄) with the equivalence ratio ranges from 0.8 to 1.2 and wall temperature 289 K to 423 K. They concluded that the heat transfer was independent of the equivalence ratio and depends on the wall temperature.

Catalytic surface reaction can be hypothesized as partial or total, liberation of reactive intermediate species as well as heat. Both these can lead to activation of the adjacent combustible mixture. As a result of catalytic pre-reaction, the required ignition energy is reduced and the flame velocity is increased (Ramesh babu et al., 1992)..

2. Mechanism of Catalytic Combustion

There are number of researchers who made detailed analysis in catalytic combustion. The general

mechanism of catalytic combustion proposed by R L Jones (1996) is depicted in Fig. 1 At temperatures below the catalytic ignition temperature (T_{cat}), no reaction occurs, even though LPG ($CH_3CH_2CH_3$ or C_3H_8) and oxygen are observed in the catalytic surface. When T equal to T_{cat} , reaction commences on the catalytic surface, with highly reactive free radical intermediates such as $CH_3CH_2CH_2^*$ and HO_2^* being formed, and

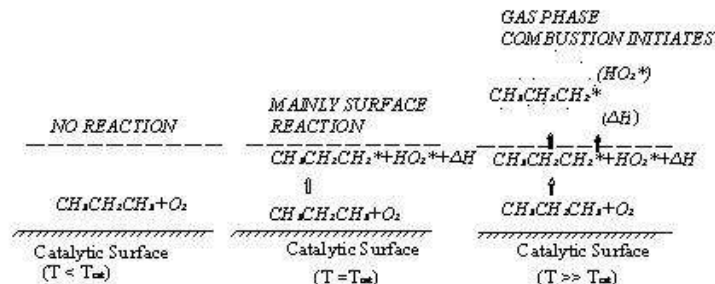
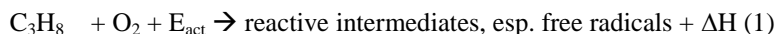


Figure 1 Regimens of surface catalytic combustion as a function of surface temperature

heat (ΔH) released, within the boundary layer above the catalytic surface. At $T \gg T_{cat}$, the reactive intermediates and heat generated at the catalytic surface pass beyond the gas/surface boundary layer and act to aid initiation of homogeneous gas-phase combustion within the bulk gas.

The combustion of LPG in air was chosen for the present study because LPG is recognized in combustion science as being the lightest straight-chain hydrocarbon whose oxidation reactions are typical of higher molecular weight hydrocarbon fuels (Hautman et al. 1981). In the combustion of LPG in air, the overall reaction is



For homogeneous combustion, an activation energy, E_{act} , of about 25-50 kcal/mol is required. The combustion mixture must be heated therefore for reaction (1) to proceed, with the spontaneous ignition temperature for LPG in air being given as 493 °, although this depends on conditions and is not a true constant. Note also that the combustion of even a simple gas such as LPG in reaction (1) actually very complex, and may involve as many as 100 intermediate species and 2000 interrelated reactions. When a catalytic surface is present, the activation energy for reaction (1) is lowered to as little as 11-15 kcal./mol, and saturated hydrocarbons such as C_3H_8 can be ignited in air over platinum catalyst at temperatures as low as 225 °C. Gases are flammable only within a specific domain of concentrations, situated between a lower explosion limit (LEL), below which the mixture is too lean in fuel to sustain combustion, and an upper explosion limit (UEL), above which the mixture is too lean in oxygen.

LPG and other gases fuels have common properties that provide them some advantages and disadvantages relative to gasoline. Before discussing its usability for this experiment, its properties are compared with gasoline. Propane has lower density and stoichiometric air-fuel ratio than gasoline and thus it could reduce the specific fuel consumption and exhaust emissions. If a propane fueled SI engine operates at the same equivalence ratio as similar gasoline fueled engine, higher power could be expected due to higher calorific value of propane. However these advantages may be balanced by decreasing volumetric efficiency. On the other hand, propane can be used at higher compression ratios due to its higher octane number, and as a consequence of this property, engine performance, engine power and thermal efficiency, would be improved.

The most important drawback of this fuel is that it reduces the engine volumetric efficiency and consequently the fresh charge mass, which is mainly due to its rising temperature and its entering the intake system in the gaseous state.

Comparisons show that if LPG fueled SI engines are operated at the same conditions with those gasoline fueled SI engines, significant improvements in exhaust emissions can be achieved

Table 1 Properties of LPG and gasoline

Sl. No	Property	LPG	Gasoline
1	Molecular formula	C_3H_8	C_7H_{17}
2	Molecular weight kg/kmol	44.10	101.213
3	Density at 15 °C kg/m ³	507	690
4	Lower heating value MJ/kg	46.40	44
5	Heat of vaporization MJ/kg	0.426	0.33
6	Stoichiometric fuel-air ratio	0.0638	0.0659
7	Research octane number	113	91

3.Experimental Setup

The experimental setup is fabricated using a simple vertically mounted furnace, which consists of a 170 mm long section, 90 mm inner diameter and 100 outer diameters. At the bottom of the vessel, perforated section is provided for uniform flow of air fuel mixture. Inner side of this vessel one cylindrical tube coated with the required material is placed, whose inner diameter is 44 mm and outer diameter is 50 mm and a height of 300 mm out of which 136 mm is placed inside the cylindrical vessel. An electrical heater is placed inside the inner tube. The layout of the experimental setup is shown in fig.2. The heater is placed inside the metal tube surface, which is again placed inside the cylindrical vessel. The heater is switched on and the steady state temperature of 100 °C is maintained. The air is flows from the compressor to the cylindrical tube through an air Rota meter and through perforated holes. The temperatures at top, bottom, packed region, inlet and outlet locations were noted at every 60 seconds intervals. The fuel flow from the LPG cylinder through the fuel Rota meter. The air and fuel are mixed in the mixing chamber; this mixture is passed over the heated surface. The temperature versus time was noted for the same locations. The procedure is repeated by changing the different coated surfaces.

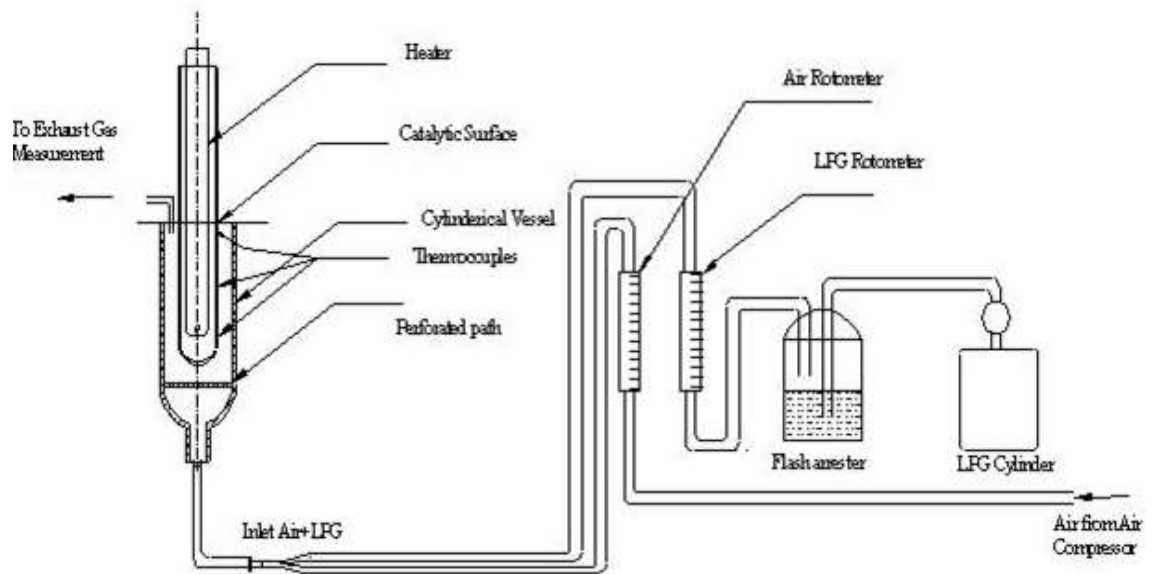


Figure 2. Layout of the experimental setup

Results and discussion.

The variation of surface temperature with time for the copper catalytic surface is shown in Fig. 3. There is no significant deviation in temperature rise for different equivalence ratio (ϕ) until the temperature reaches 300 °C, when compared to air. After this point the surface temperature rises due to catalytic reaction of the surface. The peak surface temperature was achieved, when the equivalence ratio of 1.4. This indicates that the surface reaction is more for $\phi=1.4$ than the other air fuel mixtures ($\phi=1.2$ and 1.6).

The typical surface reaction of nickel catalytic coated surface is shown in Fig. 4. The catalytic reaction for the nickel surface was activated above 270 °C, when compared with air for all the equivalence ratio other than $\phi=1.6$. The reaction was noted in the lean mixture of equivalence ratio of 1.6 at 380 °C. The maximum surface temperature was observed for equivalence ratios 1.2 and 1.4. This indicates that the surface reaction is more for $\phi=1.2$ and $\phi=1.4$, either too rich or too lean ratios gives lower peak surface temperature.

Figure 5 indicates the catalytic combustion with chromium surface for different equivalence ratios. It is noted from the figure, the temperature of the surface was raised up to 450 °C and there was no further temperature rise. This indicates the equilibrium temperature of air was attained at this point. The maximum, temperature difference was observed for $\phi=1.2$ than other air fuel ratios. While comparing Fig. 3, 4 and 5, the maximum temperature rise was noted for chromium surface with $\phi=1.2$ followed by copper with $\phi=1.4$ and nickel with $\phi=1.2$ and 1.4.

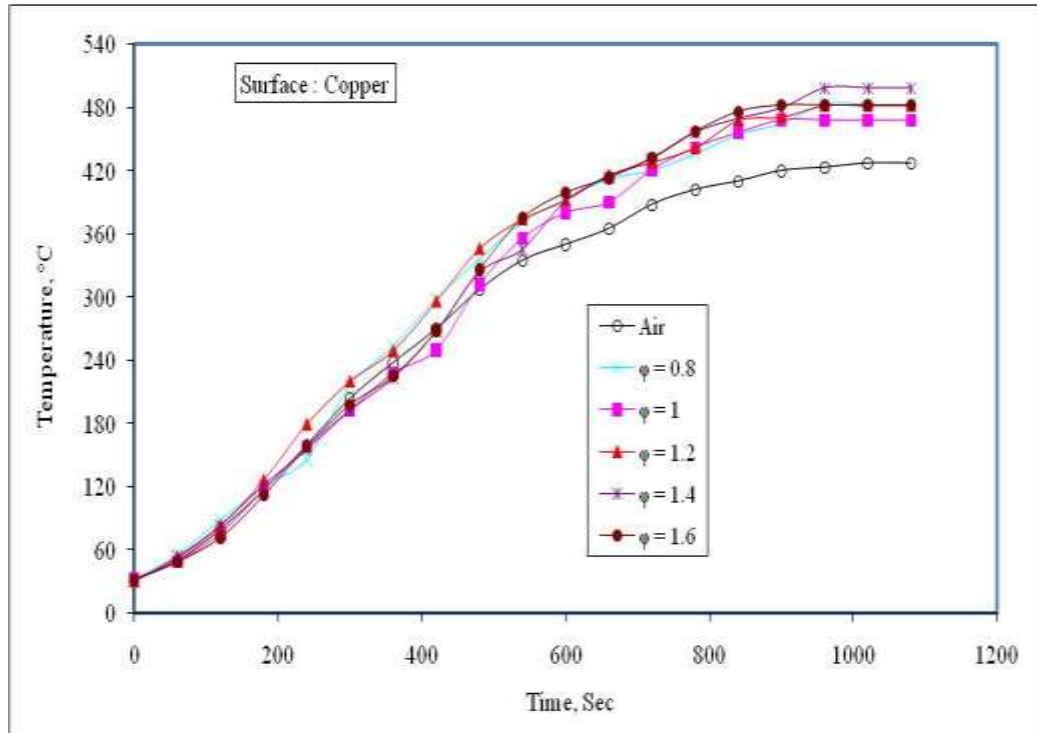


Figure 3. Variation of surface temperature with time for copper

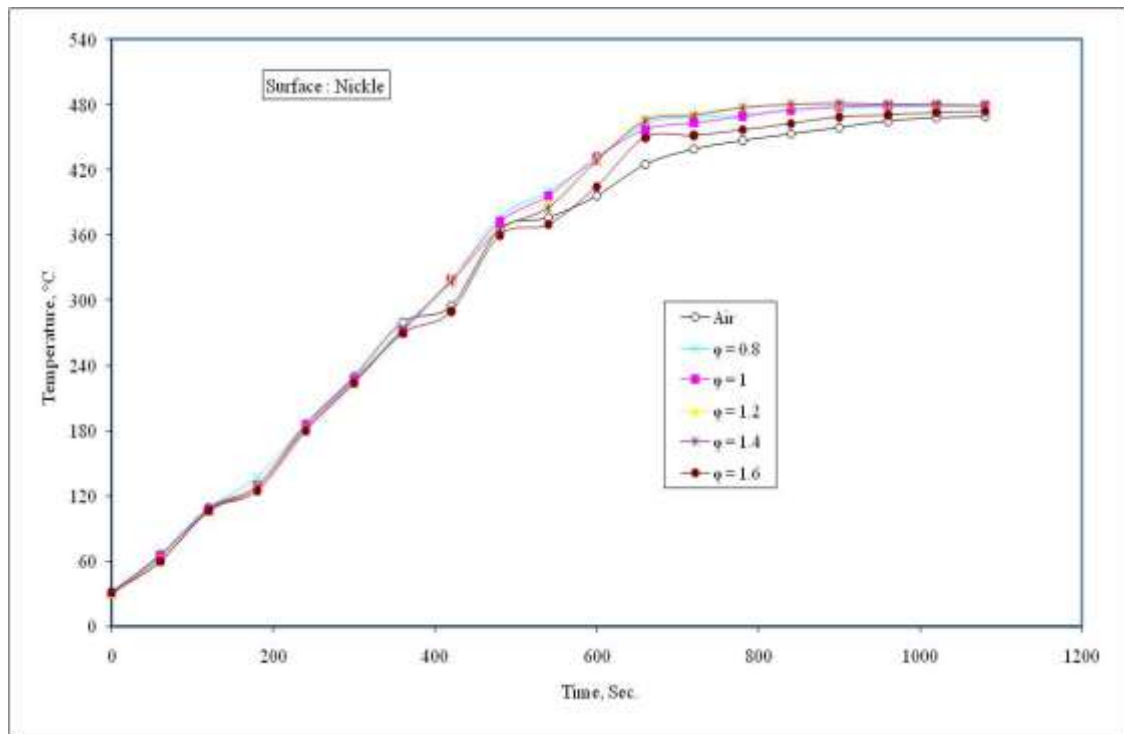


Figure 4. Variation of surface temperature with time for Nickel

The inlet temperature was measured before the gas mixture enters to the chamber and exhaust temperature was measured at the outlet of the test chamber. The differences in temperature for different equivalence ratios with copper, nickel and chromium are given in Fig. 6, 7 and 8 respectively. From the Fig. 6, the maximum temperature difference was observed in rich mixture $\phi=0.8$. However for too lean mixture $\phi=1.6$ there was no much variation in temperatures. It was observed maximum temperature difference at $\phi=0.8$ and $\phi=1$, for lean mixture the temperature difference is very small it shows in Fig.7. From the Fig. 8 the maximum temperature was observed in the $\phi=1.2$, and for $\phi=0.8$ and $\phi=1$ maximum temperature obtained compare to $\phi=1.2$, the temperature difference is very small.

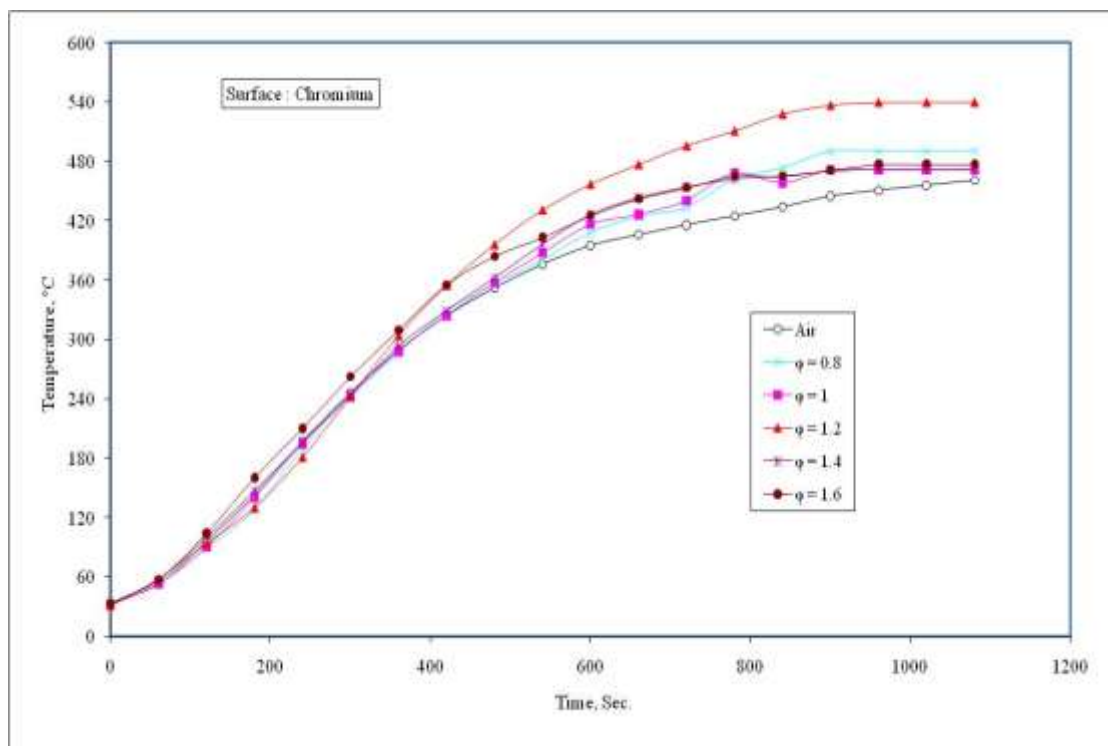


Figure 5. Variation of surface temperature with time for chromium

It was observed from the Fig. 9 for copper material at rich mixture $\phi=0.8$ $\phi=1$ and $\phi=1.2$ shows the maximum temperature difference inlet and outlet But in lean mixture $\phi=1.4$ and $\phi=1.6$ maximum temperature was observed in nickel material

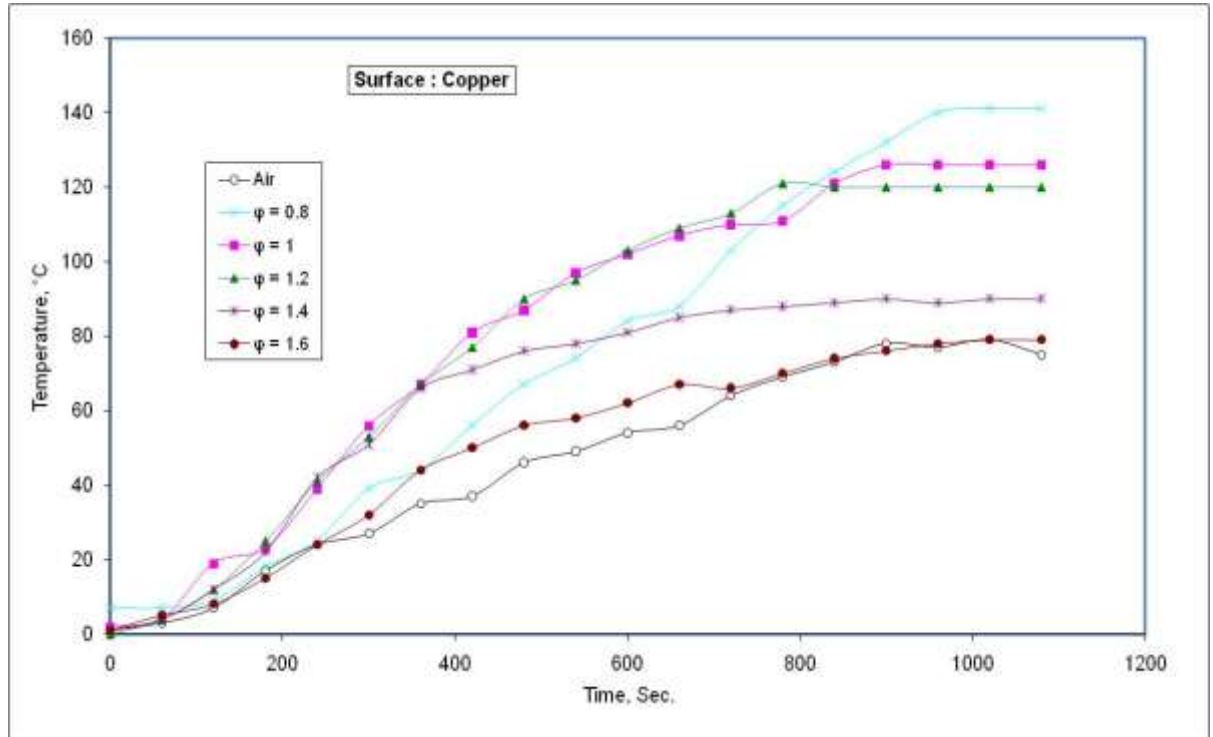


Figure 6. Variation of inlet and outlet temperature difference with time for copper

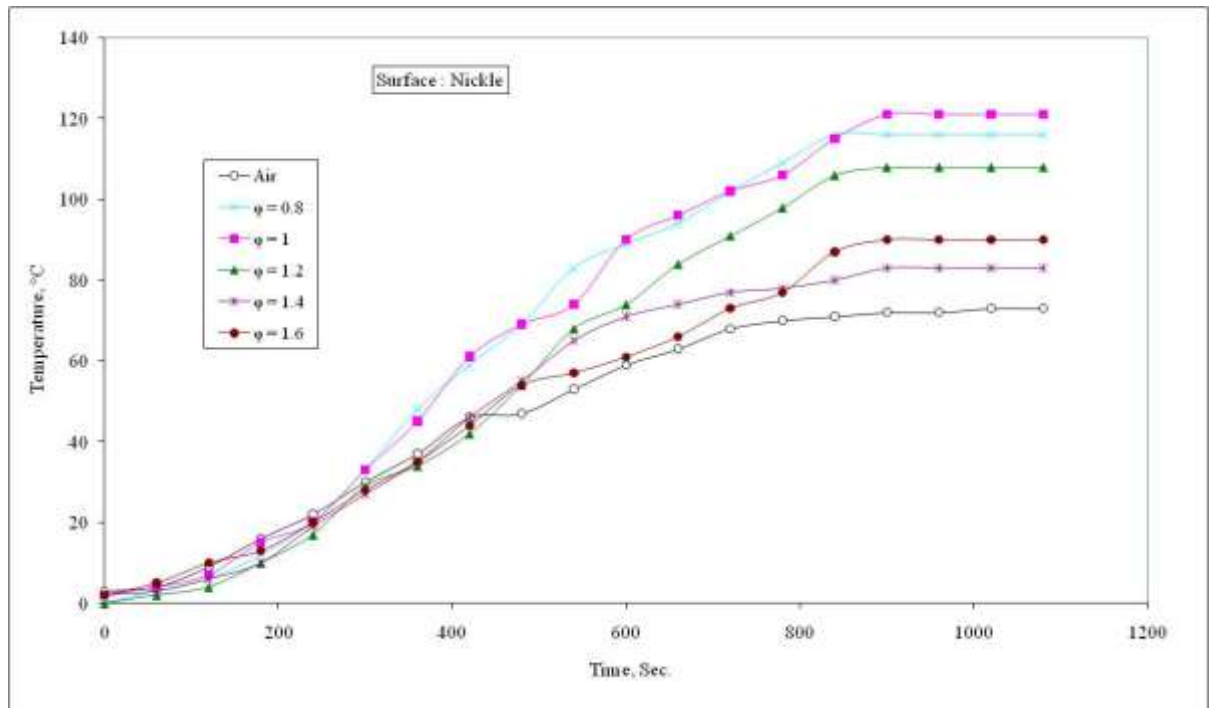


Figure 7. Variation of inlet and outlet temperature difference with time for Nickel

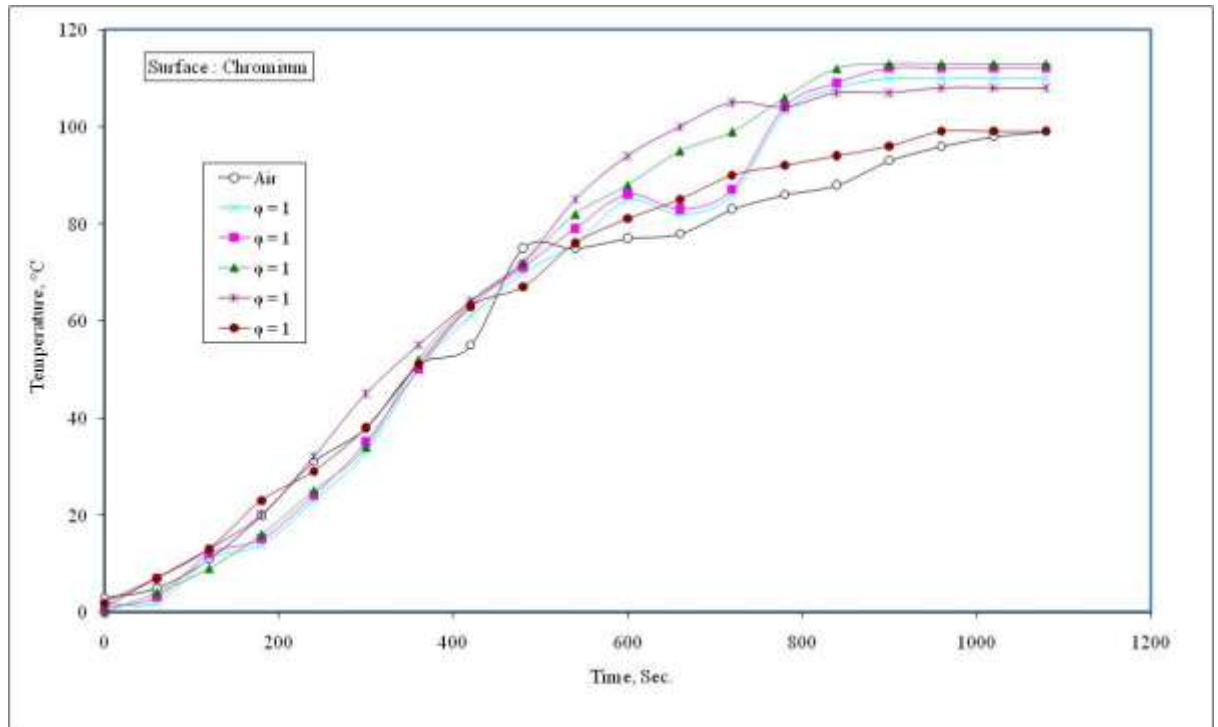


Figure 8. Variation of inlet and outlet temperature difference with time for Chromium

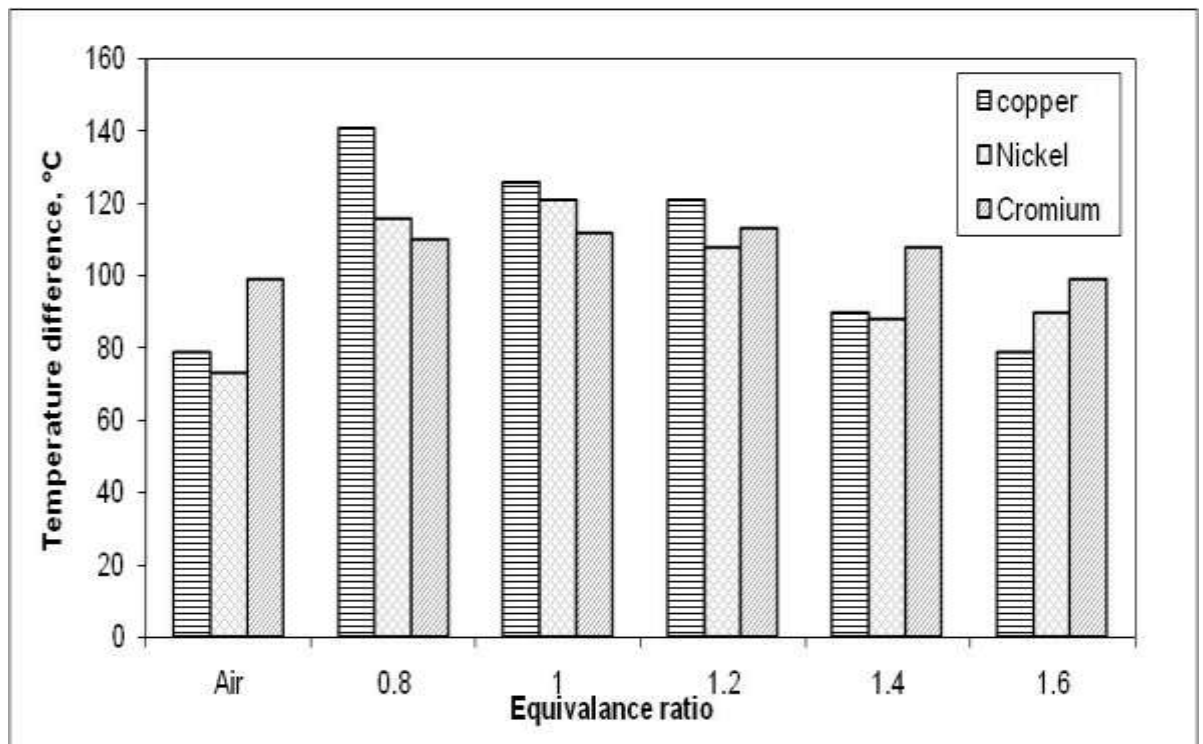


Figure 9. Comparison of maximum temperature difference with different catalytic surfaces and equivalence ratios.

5. Conclusion

From the above discussions the following conclusions are arrived.

- For copper material at equivalence ratio(ϕ) of 1.4 at the point 300°C surface temperature rises due to catalytic reaction of the surface the peak surface temperature obtained.
- The catalytic reaction for the nickel surface shows activation above 270 °C, when compared with air for all the equivalence ration other than $\phi=1.6$. The reaction was noted in the lean mixture of equivalence ratio of 1.6 at 380°C. The maximum surface temperature was observed for equivalence ratios 1.2 and 1.
- For chromium the maximum, temperature difference was observed for $\phi=1.2$ than other air fuel ratios. While comparing all the catalytic material, the maximum temperature rise was noted for chromium surface with $\phi=1.2$ followed by copper with $\phi=1.4$ and nickel with $\phi=1.2$ and 1.4.
- For copper material at rich mixture $\phi=0.8$ $\phi=1$ and $\phi=1.2$ shows the maximum temperature difference inlet and outlet But in lean mixture $\phi=1.4$ and $\phi=1.6$ maximum temperature was observed in nickel material.

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Nomenclature

E_{act} – Activation energy

CO- carbon monoxide

T_{cat} – Catalytic surface temperature

Cr – Chromium

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Cu – Copper
 ϕ - Equivalence ratio
Ni – Nickel
NO_x- Nitrogen oxides
PM - particulate matter
Ag – Silver

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