Studies of carbon monoxide oxidation at ambient conditions

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Abstract

Carbon monoxide (CO) oxidation was studied at ambient condition over hopcalite ($CuMn_2O_2$) based catalyst. The catalysts were prepared by co- precipitation method. The prepared hopcalite based catalysts, Cu-Mn-Co catalyst, Cu-Mn-Cu catalyst, Cu-Mn-Ce catalyst were examined for oxidation of CO in fixed bed tubular flow reactor. The CO is generally present at lower temperature, as it is a result of the incomplete combustion of hydrocarbons. By observation it was found that the amount of Carbon dioxide was increasing with temperature in outlet gases. Among various catalysts studied, Cerium promoted Hopcalite showed maximum catalytic activity for CO oxidation at ambient condition. The conversion of CO over Cu-Mn-Ce was started at room temperature and gives 100% CO conversion in 75 minutes. Thus, it can be concluded that the copper based catalyst would be an attractive alternative to presently used noble metals catalyst for oxidation of CO at ambient condition. The prepared hopcalite based catalysts, are low cost and resistant to moisture.

Keywords: CO, ambient conditions, hopcalite.

INTRODUCTION

CO can prove perilous without giving any physical indication because it is a tasteless, odourless and colourless gas. It is chemically inert under normal conditions with estimated atmospheric mean life of about 2.5 months [1]. Vehicular exhaust contributes about 64% CO pollution in developed countries [2]. This can seriously affect human aerobic metabolism because of its affinity to haemoglobin being 210 times greater than that of oxygen and so, if its concentration becomes higher than 0.0998% by volume in atmosphere, it can lead to the failure of the process of combination of oxygen with the haemoglobin resulting ultimately in respiratory failure and consequent death. It forms a very stable compound called carboxyhaemoglobin (COHb) in blood whose 1-2% concentration can cause headache, fatigue, drowsiness and may have effect on human behavioural performance, 2-5% concentration may cause impairment of time interval discrimination, visual acuity, brightness discrimination and certain other psychomotor functions which may lead to accident on roads. 5-10% concentration can lead to changes in cardiac and pulmonary functions and 10-80% can cause coma, respiratory failure and death [3]. CO has detrimental effect on vegetation. It CO has detrimental effect on vegetation. It affects N_2 fixation ability of bacteria, causes leaf curling, reduction in leaf size and chlorophyll with premature ageing etc. It also inhibits cellular respiration in plants by 3 reacting with the enzyme system cytochrome oxidase [4]. Thus it affects agricultural productivity. CO is only a very weak direct greenhouse gas, but has important indirect effects on global warming. Carbon monoxide reacts with hydroxyl (OH) radicals in the atmosphere, reducing their abundance. As OH radicals help to reduce the lifetimes of strong greenhouse gases, like methane, carbon monoxide indirectly increases the global warming potential of these gases [5].

The catalytic oxidation of CO with the object of reducing air pollution is actually an important consideration when one thinks in terms of automobile emission control at low temperature [6]. The high cost of noble metals and their low availability, makes it a compulsion to consider transition metal catalyst as an alternative [7].

EXPERIMENTAL
Catalyst preparation

Four hopcalite catalysts were prepared by co-precipitation method. The catalysts prepared by this method contain copper, maganese, cobalt, ceria and silver for ambient condition CO oxidation.

Hopcalite (Cu-Mn) catalyst

Catalyst was prepared using the co-precipitation method. Aqueous solutions of $Cu(NO_3)_2 \cdot 3H_2O$(6.4g/100ml) and $Mn(NO_3)_2 \cdot 6H_2O$ (4.48g/100ml) were pre-mixed in a 2:1 ratio. The resulting solution was stirred and heated to 80 °C and fed using a peristaltic pump to a stirred precipitation vessel maintained at 80 °C. Simultaneously, an aqueous solution of Na_2CO_3 (21.2g/100ml) was added via a burette to maintain the pH at 8.3. The precipitate was immediately recovered by filtration, washed several times with hot distilled water, dried in air (120 °C for 16 h) and calcined in static air at 415 °C. This catalyst is named as cat-A.

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Cu-Mn-Co catalyst

This catalyst prepared using a coprecipition method. The a Aqueous solutions of $Cu(NO_3)_2 \cdot 3H_2O$ (5.8636g/100ml), $Mn(NO_3)_2 \cdot 6H_2O$ (10ml/100ml) and $Co(NO_3)_2.3H_2O$ (0.2124g/100ml) were mixed . The resulting solution was stirred and heated to 80 °C and fed using a peristaltic pump to a stirred precipitation vessel maintained at 80 °C. Simultaneously, an aqueous solution of Na_2CO_3 (21.2g/100ml) was added via a burette to maintain the pH at 8.3. The precipitate was immediately recovered by filtration,washed several times with hot distilled water, dried in air (120 °C for 16 h) and calcined in static air at 415 °C. This being the main variable investigated in this study. Cu-Mn-Co catalyst is named as cat- B [8].

Cu-Mn-Ce catalyst

This catalyst prepared using a co-precipition method. The a Aqueous solutions of $Cu(NO_3)_2 \cdot 3H_2O$ (5.8636g/100ml), $Mn(NO_3)_2 \cdot 6H_2O$ (10ml/100ml) and $Ce(NO_3)_2. 3H_2O$ (0.316987g/100ml) were mixed . The resulting solution was stirred and heated to 80 °C and fed using a peristaltic pump to a stirred precipitation vessel maintained at 80 °C. Simultaneously, an aqueous solution of Na_2CO_3 (21.2g/100ml) was added via a burette to maintain the pH at 8.3. The precipitate was immediately recovered by filtration, washed several times with hot distilled water, dried in air (120 °C for 16 h) and calcined in static air at 415 °C. This being the main variable in- vestigated in this study. Cu-Mn-Co catalyst is named as cat-C.

Cu-Mn-Ag catalyst

This catalyst prepared using a coprecipition method. The a Aqueous solutions of $Cu(NO_3)_2 \cdot 3H_2O$ (5.8636g/100ml), $Mn(NO_3)_2 \cdot 6H_2O$ (10ml/100ml) and $Ag(NO_3)_2.3H_2O$ (0.123954g/100ml) were mixed . The resulting solution was stirred and heated to 80 °C and fed using a peristaltic pump to a stirred precipitation vessel maintained at 80 °C. Simultaneously, an aqueous solution of Na_2CO_3 (21.2g/100ml) was added via a burette to maintain the pH at 8.3. The precipitate was immediately recovered by filtration,washed several times with hot distilledwater, dried in air (120 °C for 16 h) and calcined in static air at 415 °C. This being the main variable investigated in this study. Cu-Mn-Ag catalyst is named as cat-D.

Designation of Catalyst

The designation and elemental compositions of the reduced catalysts are A, B, C, and D. The symbols showing the catalyst name and their details are given below:

Table 1. Types of catalyst prepared and their conditions.

S.No.	Catalyst Name	Catalyst composition	Calcination Temperature(°C)
1	A	Hopcalite	415°c
2	B	Cu-Mn-Co	415°c
3	C	Cu-Mn-Ce	415°c
4	D	Cu-Mn-Ag	415°c

Reaction Condition and flow rate:

One ml (0.7839g) of catalyst was placed in the reactor over glass wool previously packed in the reactor. The constant flow rate of air from cylinder was maintained in the reactor. The CO flow rate was maintained at desired level by using the syringe pump. The CO:air ratio was maintained 1:199 throughout the experiments. The reaction products were analyzed by Gas Chromatography. The conversion is calculated by knowing the inlet and outlet concentration of CO using the following formula:

$$X_{CO} = (C_{COin} - C_{COout})/ C_{Coin}$$

RESULT AND DISCUSSION

The conversion of CO over cat-A was started at room temperature which increases with time and after sometime it decreases as shown in figure 1. The maximum conversion achieved by this catalyst was 33% at 48 °C.
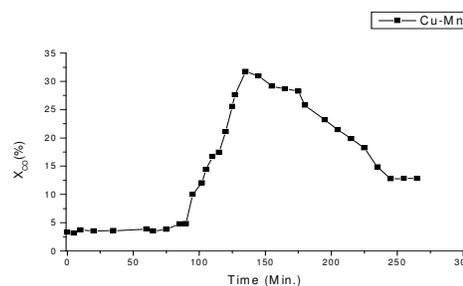

Fig 1. Percent total conversion of CO at various time with catalyst A at temp. 48 °C
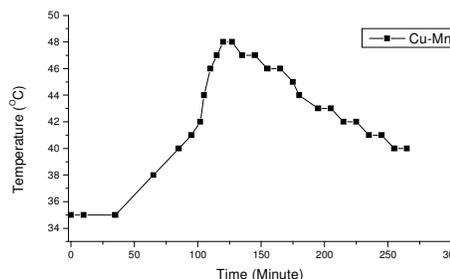

Fig 2. Percent total conversion of CO at various temperatures with catalyst A.

The reaction being exothermic causes temperature of the catalyst bed to increase itself with time and after sometime it decreases showing a maxima as shown in figure 2.
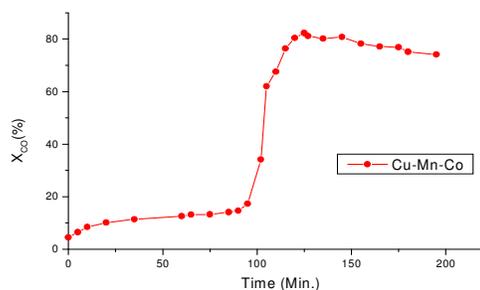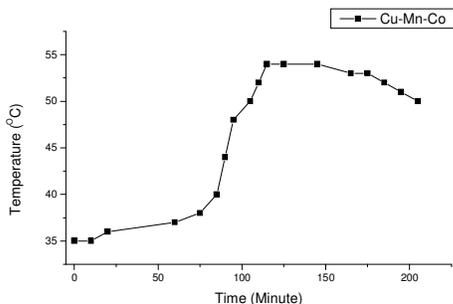

Fig 3. Percent total conversion of CO at various times with catalyst B at temp.54°C.

The conversion of CO over cat-B started at room temperature which increases with time and after sometime it decreases as shown in figure 3. The maximum conversion achieved by this catalyst is 80% at 54°C.

The reaction being exothermic causes temperature of the catalyst bed to increase itself with time and after sometime it decreases showing a maxima as shown in figure 4.

Fig 4. Percent total conversion of CO at various temperatures with catalyst B.

The conversion of CO over cat-C was started at room temperature which increases with time and after sometime it decreases as shown in figure 5. The maximum conversion achieved by this catalyst is 100% at 65°C.
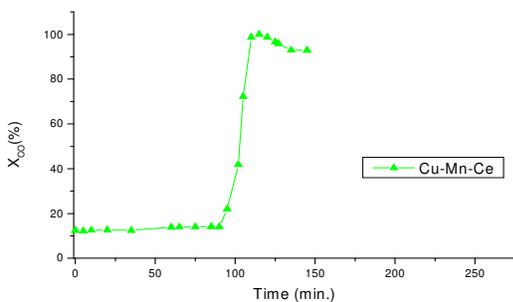

Fig 5. Percent total conversion of CO at various times with catalyst C at temp.65°C.

The reaction being exothermic causes temperature of the catalyst bed to increase itself with time and after sometime it decreases showing a maxima as shown in figure 6.
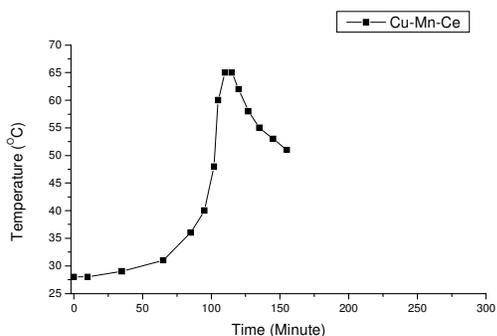

Fig 6. Percent total conversion of CO at various temperatures with catalyst C.

The conversion of CO over cat-D was started at room temperature which increases with time and after sometime it decreases as shown in figure 7. The maximum conversion achieved by this catalyst is 56% at 47°C.
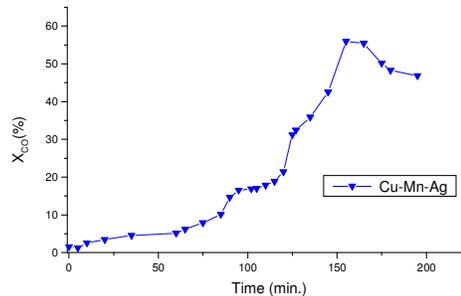

Fig 7. Percent total conversion of CO at various time with catalyst D at temp. 47°C.

The reaction being exothermic causes temperature of the catalyst bed to increase itself with time and after sometime it decreases showing a maxima as shown in figure 8.
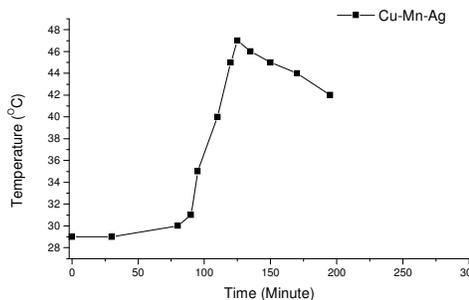

Fig 8. Percent total conversion of CO at various temperatures with catalyst
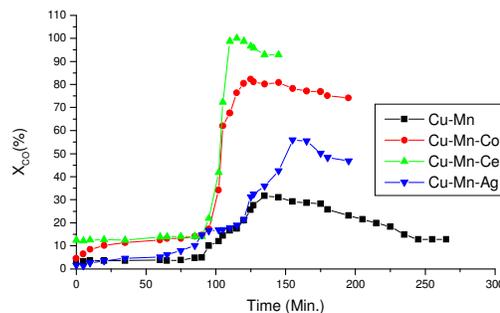

Fig 9. Percent total conversion of CO with time over different catalysts.

Figure 1 to 8 shows the activity in terms of conversion of individual catalyst and Figure 9 showing the activity comparison for all catalyst.

As curves are showing with all catalysts, there is no CO_2 found in product gases below 31 °C temperature. At the temperature above the 31°C, catalysts starts showing some activity but still the amount of products (CO_2) was less. The CO is generally present at lower temperature, as it is a result of the incomplete combustion of hydrocarbons. By observation it is found that the amount of CO_2 is increasing with temperature in outlet gases. Among various catalysts studied, Cerium promoted Hopcalite shows maximum catalytic activity for CO oxidation at ambient condition. The conversion of CO over Cu-Mn-Ce, cat-C was started at room temperature and gives 100% CO conversion in 75 minutes.

CONCLUSION

Thus, it can be concluded that the copper based catalyst would be an attractive alternative to presently used noble metals catalyst for oxidation of CO at ambient condition. The activity order of various studied catalysts can be arranged in the following order: cat-C > cat-B > cat-D > cat-A. The catalyst developed can be very effective to remove CO from the breathing air by incorporating it into gas mask or breathing apparatus.

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