

# NON ADIABATIC CATALYTIC COMBUSTION OF CARBON MONOXIDE USING FLOW REVERSAL

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## Abstract

*An experimental study of non-adiabatic fixed bed reactor for controlling CO gas has been made under flow reversal operation. Temperature measurements have been made within the catalyst and packing beds. The influence of the operating parameters upon the shape of temperature profiles, conversion and heat removal were studied. The experiments showed that the maximum measured reactor temperature and heat removal are rather insensitive to changes in cycle duration; sensitivity was shown to the feed flow rate and reactant concentration.*

## INTRODUCTION

Flow reversal technology is a relatively new technique for handling exothermic reaction in fixed bed reactors. The idea of periodic reversal of the flow direction comes from Russian researchers (Boreskov et al., 1977). They used simulation to explore the concept based on a one-dimensional unsteady state mathematical model. After the pioneering investigation of Boreskov et al. (1977), many researchers have worked experimentally and analytically on flow reversal (Boreskov et al., 1982; Eigenberger and Nieken, 1988; Gawdzik and Rakowski, 1989; Matros, 1989 and 1990; Blank et al., 1990; Bathia, 1991; Gupta and Bathia, 1991; and Rehacek et al., 1992). There have been also industrial scale plants (Matros, 1989 and 1990).

Matros (1989 and 1990) studied the possibility of using flow reversal technology, analytically and experimentally, for the purification of gas-air mixture containing small amounts of carbon monoxide, methane or other combustible substances which are formed during various processes. Several researchers have worked using the flow reversal technology in the process other than SO<sub>2</sub> oxidation and purification of combustible gases. Blank et al. (1990) proposed to use this technique for production of synthesis gas, carbon monoxide and hydrogen, from catalytic partial oxidation of natural gas with air. Froment (1990) reported research on the operation of a reversal flow reactor for a low pressure methanol synthesis. In 1992, Haynes et al. proposed the application of reverse flow reactor to endothermic reactions for the dehydrogenation of ethyl benzene.

Among these and the previous studies (Boreskov et al., 1977 and 1983; Matros, 1989 and 1990), there is no published work on the handling of runaway when the

feed concentration suddenly increases. So far the flow reversal process is carried out using two adiabatic reactors connected together with a heat exchanger placed between them. In this kind of process, it is difficult to control runaway. Therefore, we propose to improve the technique of handling low gas concentrations by integrating a fixed bed reactor with a heat exchanger for removing heat directly from the system.

## EXPERIMENTAL AND PROCEDURE

### Equipment and procedure

Figure 1 is a schematic diagram of the experimental set-up. All gases flowing to the reactor were dried over filters containing 4A molecular sieves and anhydrous calcium sulphate to reduce traces of impurities. Brooks 5850 flow transducer-controllers regulated the feed mass flow rate into the reactor and the heat exchanger when the air from the system was used as a coolant. They permitted only uni-directional flow and therefore acted as check valves. This feature insured that during flow reversal, for instance, there was no backflow immediately after switching. The three ways Tomco 95 solenoid valves controlled by a ChronTrol CD 03 timer were used for flow reversal operation when particular feed mixtures were fed into the system at preset times. The system operating pressure was measured by Matheson pressure gauges. All the thermocouples in the system were connected to an Instrulab 2000 data logger. Effluent concentrations of the carbon monoxide and carbon dioxide formed from the reaction were measured by an on-line IR spectrophotometer IR-702 D dual analyser, supplied by Infrared industries, Inc. This unit was capable of

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continuously monitoring CO and CO<sub>2</sub> concentrations in the product stream simultaneously.

The reactor was 15 cm long and 1 cm ID stainless steel tubing with a water cooler surrounding it and connected to a 30-cm length of packing which had 1.6-cm ID as a preheater at each end. To prevent catalyst packing ball movement, stainless steel screens were installed at the inlet and outlet of the reactor.

Nine 1.6-mm OD chromel-alumel thermocouples were placed in each packing zone and seven 0.508-mm OD chromel-alumel thermocouples were inserted in the reaction zone to monitor the temperature distribution as reaction proceeded. To make sure that the thermocouples only read the gas temperature, they were jacketed using ceramic thermocouple protection tubes.

Four 1.6-mm OD chromel-alumel thermocouples were placed in the cooling system. Two of them were used to measure the inlet and outlet coolant temperatures. The other two were used to measure the reactor wall temperatures. These four thermocouples were not jacketed.

For each run, the reactor was firstly preheated to slightly above desired reaction temperature using a heating tape wrapped along the gas tubing. To make sure that the temperature in the system was uniform, preheated hot air was flow to the system. When a constant temperature was reached, the heating tape was turned off and the cold reaction mixture was introduced into the system. As this mixture passed through the hot packing, it absorbed heat. As the packing cooled, the mixture was heated to the reaction temperature and reacted over the catalyst. The exothermicity thus released raised the temperature of the catalyst bed. Heat was carried in the hot gas to the second packing zone and formed a slowly migrating front moving towards the outlet in that zone. After some time, the region in the inlet of the reactor would cool as heat transferred from the packing to the feed gas. Meanwhile, the outlet region was being heated by the exothermic reaction. The direction of the gas mixture was then reversed, and the reaction wave started to creep in the opposite direction in the two packing zones. After some time the whole cycle would be repeated. This cyclical process created a hot region in the catalyst bed which could be used as a heater or boiler. The temperature profiles along the reactor were monitored and recorded in a data logger during the process. The monitoring and recording of temperatures were started when the cold reactant was first introduced to the reactor. The carbon monoxide and carbon dioxide concentration of the gas leaving the system were monitored continuously with an on-line analyzer. The monitoring of the concentration was also started when the cold reactant was first introduced. Figure 2 shows the schematic diagram of flow reversal process.

To avoid excessive heat loss, another heating tape was placed in the outermost layer of the insulation. This heating tape was controlled so as to have the same temperature as the initial temperature of the system.

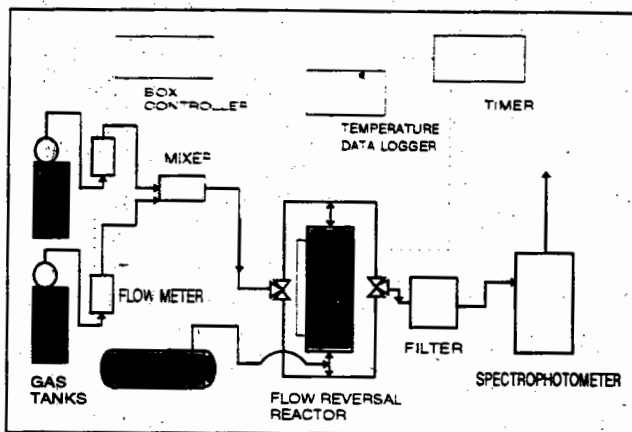


Figure 1. Schematic diagram of the experimental set-up.

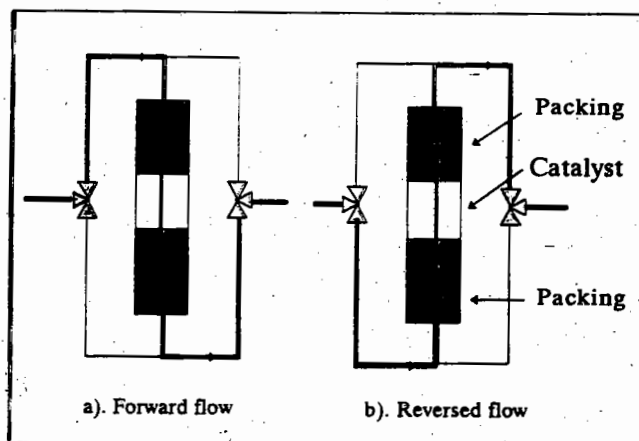


Figure 2. Flow reversal process.

## Materials

Pellets of commercial catalyst, platinum on gamma alumina were used in the process. The Pt/g-Al<sub>2</sub>O<sub>3</sub> catalyst contained 0.1 % Pt (Girdler T-309), the platinum being deposited as a shell surrounding the catalyst particle supplied as 3.18 x 10<sup>-3</sup> m time 3.18 x 10<sup>-3</sup> m tablet. The physical properties of the catalyst pellets are given in Table 1. The diameter of balls of chrome steel alloy used as packing to retain heat is 0.32 cm.

Table 1. Catalyst properties

Surface area	2.01 x 10 <sup>5</sup> m <sup>2</sup> /kg
Bulk density	1.8 x 10 <sup>3</sup> kg/m <sup>3</sup>
Total pore volume	2.4 x 10 <sup>-4</sup> m <sup>3</sup> /kg

In this research, 99% purity of carbon monoxide was used as feedstock. CO was chosen because its oxidation can be initiated at relatively low temperatures ( $\gg 200^\circ\text{C}$ ). It is important industrial waste gas. Catalytic oxidation is often used to render this gas inert by converting it to  $\text{CO}_2$ .

## RESULTS AND DISCUSSION OF STATIONARY CYCLING STATE

### Preliminary experiment

To make sure that the results would not be unintended artifacts of the experimental system, a preliminary test was carried out prior to the flow reversal experiments. The materials of construction of the reactor (stainless steel), packing materials (chrome-alloy balls), thermocouples and thermocouple cement used for cementing thermocouples were tested for a possible catalytic effect. This was done by passing the reactant mixture through the heated reactor tube without catalyst. In these experiments, no conversion was detected.

The experimental results reported below show several interesting features of the reaction system such as the occurrence of a maximum temperature close to the reaction zone entrance and the occurrence of the second maximum in the first minute when the flow is reversed. These features can be understood by considering the intrinsic kinetics, the high exothermicity of the reaction and thermal inertia. Explanation have been offered in the literature and these will be discussed in further detail in this section.

### Temperature profile

The results of the experiments are shown in Figures 3 to 12. The responses sought were the maximum temperature in the bed, conversion of the combustant and the amount of heat that could be removed. The maximum temperature is actually the maximum thermocouple reading. This is an estimate of the maximum temperature and certainly less than the maximum temperature. Because of the small diameter of the reactor, it was not possible to monitor the temperature profiles in the radial direction. Therefore the temperature profiles presented in this study are only in the axial direction. All experiments were done for 15 cycles. The number of cycles chosen was based on the experiment exhibiting the maximum temperature. The experiments show that after 7 cycles the maximum temperatures for different level of variables are constant as shown in Figure 3 and we assumed that by this time the system had become stable. However, to make sure that we worked in a stable region, the number of cycles chosen was 15. From the factorial experiments, a "base case" was chosen as: CO concentration = 5%, feed flow rate = 500 ml/min, initial temperature =  $200^\circ\text{C}$  and the cycle time = 30 min. This "base case" is used as the base from which to examine the effect of each variable. Figures 4 and 5 show an example of the temperature

distributions for the base case at the end of 15<sup>th</sup> cycle, and the transient temperature profile in the reaction zone in the 15<sup>th</sup> cycle. The air coolant flow rate was 1000 ml/min, and the direction of the flow was set countercurrent to the direction of the reactant flow, for both forward and reversed flows. With this minimum set of variables and a 1000-ml/min coolant flow rate, the maximum temperature obtained in the bed was  $278.8^\circ\text{C}$  for CO oxidation at 100 % conversion. The heat removal from the system was 33.74 J/min for CO oxidation.

The axial temperature profiles in the reaction zone, i.e., the transient temperature distribution in the zone, at the end of the 15<sup>th</sup> cycle indicate the formation of a maximum temperature region, known as a hot spot, close to the reaction zone entrance. Also transient profiles show two maxima in the first minute of operation. These phenomena can be explained as follows: as the hot feed mixture enters the reaction zone, the reaction generates heat, thus elevating the catalyst surface temperature and increasing the reaction rate. Since conversion is zero at the entrance the reaction rate is high, even greater amounts of reactants are rapidly consumed and greater amounts of heat are generated. Then, as the combustant concentration downstream decreases, the reaction rate also decreases and so does the heat generated. Since the system is a non-adiabatic, some heat from the reactor is also transferred to the coolant in the radial direction; heat loss sets up an axial temperature gradient causing heat transfer in the axial direction. This results in a decrease in temperature. Thus, a maximum temperature is produced near the reaction zone inlet. The second maximum in the first minute of the operation is simply caused by thermal inertia, that is the hot spot from the previous half-cycle where the position of the second maximum is the maximum temperature location from the previous half-cycle. The second maximum disappears quickly since there is essentially no exothermicity in this region to maintain the heat.

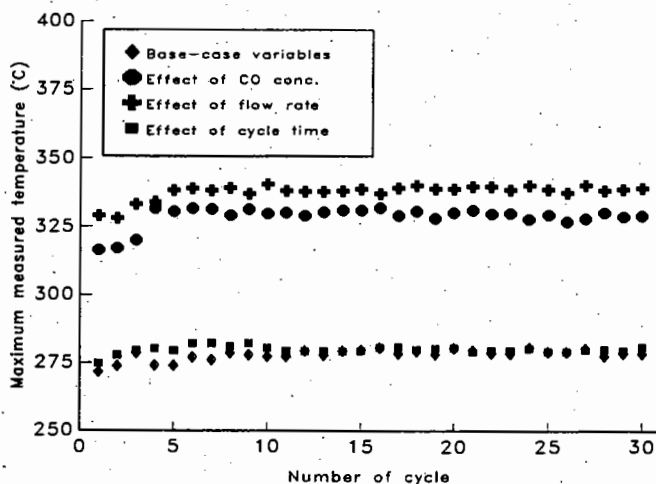


Figure 3. Effect of cycle number on the maximum temperature

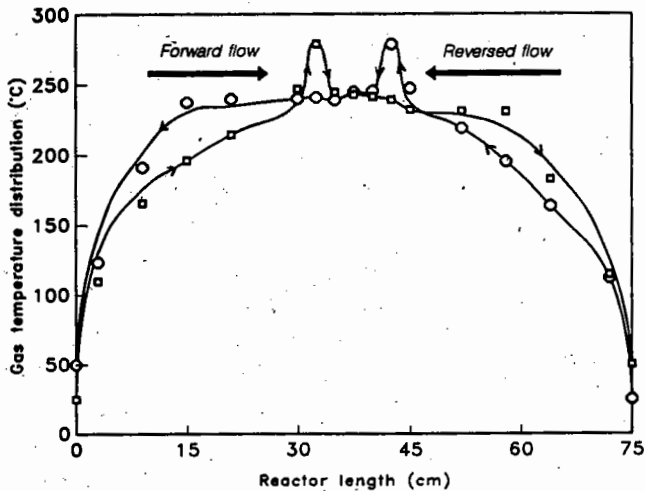


Figure 4. Temperature distribution for base-case

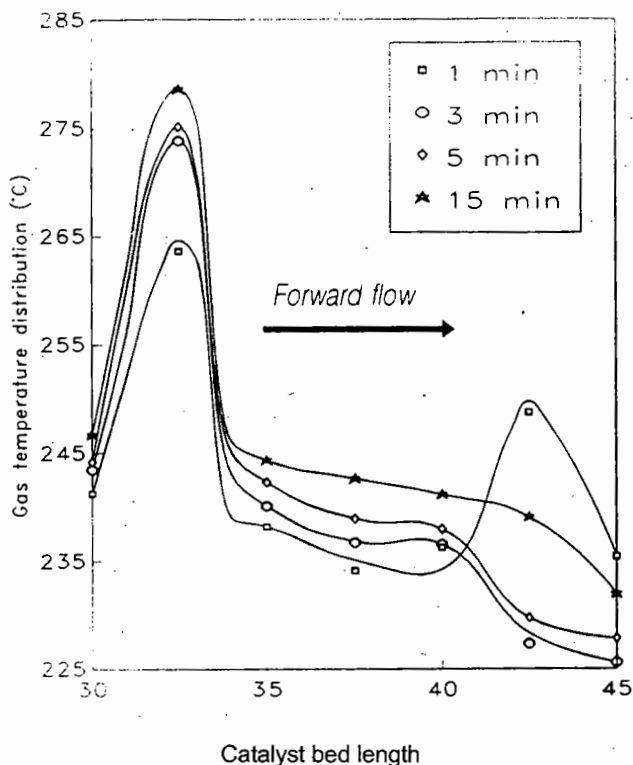


Figure 5. Temperature distribution in the reaction zone

#### Effect of concentration

The effect of CO concentration was studied by increasing the CO concentration from 5 to 10 % while keeping the other variables at their base levels. The flow rate of air coolant used was 1000 ml/min and the direction of the coolant was always countercurrent to the direction of the feed mixture inside the reactor as in the previous experiment. The effect of CO concentrations on the maximum measured temperature with other levels of variables is shown in Figures 6 and 7. From the figures, it can be seen that as the combustant concentration is increased, the maximum

measured temperature and heat removal also increase. These increases are caused by the increased of the heat released. By increasing the CO concentration from 5 to 10 %, heat released is doubled. This accounts for the change in the maximum temperature measured by 50.5 °C for CO oxidation. Despite the increased concentration, the heat removal rate increased only by 5.9 J/min for CO and 12.7 J/min for propylene. This indicate the considerable heat loss (284 J/min, from the heat balance calculation). Figure 6 and 7 show the increasing maximum measured temperature and heat removal vs concentration. The temperature profiles for different concentration of CO in the reaction zone for forward flow at the end of 15<sup>th</sup> cycle are displayed in Figure 8.

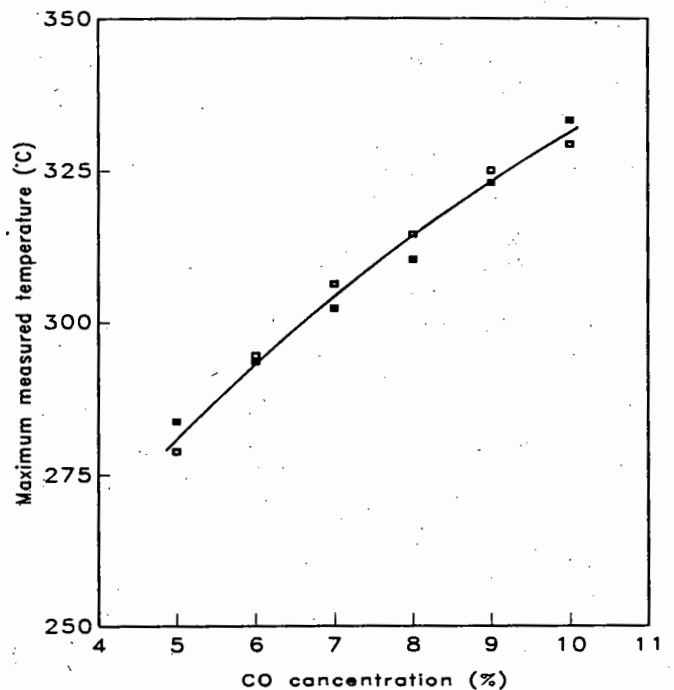


Figure 6. Effect of CO concentration on maximum temperature

Further experimentation on the variation of CO concentration shows that the measured maximum temperature and heat removal is not linear in the CO concentration. Further studies show that when the concentration is increased, not only does the maximum temperature increase but the inlet temperature and the hot region also increase which is shown by the greater area beneath the curves. As explained earlier, this is a heat release effect. However, the region at the end of the reactor is almost the same for several different concentrations for both forward and reversed flows. This result suggests that combustion goes to completion in the front of the bed so that higher heat loss at higher concentration drops the temperature faster so that at the end of catalyst bed, the outlet temperature is just a small function of temperature. Therefore the heat transferred to the packing region is not much different for either low or high concentrations.

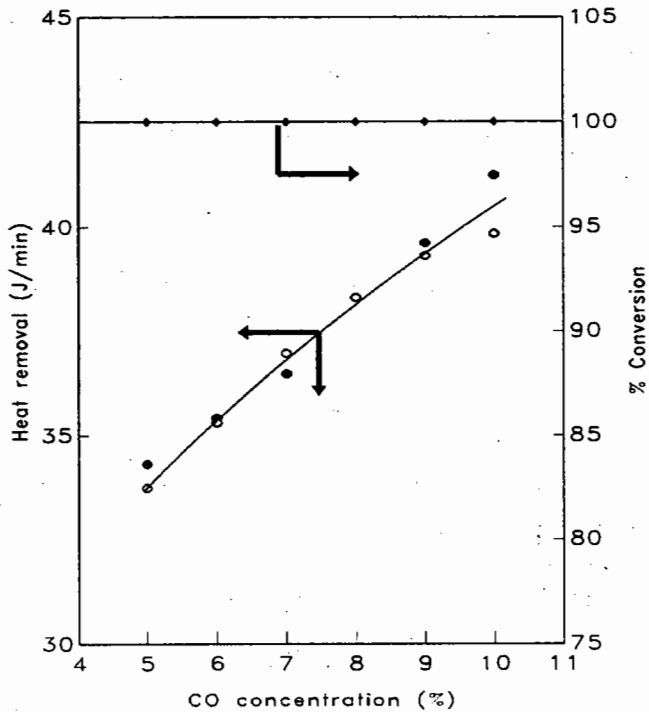


Figure 7. Effect of CO concentration on heat removal and conversion

The ratio of the maximum measured temperature rise to the adiabatic temperature rise, calculated as  $(T_{max} - T_{in}) / T_{ad}$ , and the percentage of the heat removal by the coolant from the total heat released by the reaction have also been studied as functions of the CO concentration in the feed. The result shows that the higher the concentration of CO, the lower the ratio of maximum temperature rise to the adiabatic value and the lower the percentage of heat release that is removed via coolant. These phenomena are caused by the heat loss in the radial direction from the concentric heat exchanger as a result of insufficient insulation as well as heat storage in the insulation. Higher concentrations produce higher maximum temperatures which create larger temperature differences between the system and the surroundings, thereby causing greater heat losses. This situation has also been observed by Matros (1989) in an adiabatic flow reversal reactor for methane combustion. He shows in his experiments that by increasing the concentration of methane from 0.48 to 0.66 %, the heat recovery decreases from 51 to 34 %. The heat loss for this experiment is estimated as follows:

$$Q_{\text{release}} = Q_{\text{exhaust}} + Q_{\text{coolant}} + Q_{\text{heat loss}} \quad 1)$$

where

$$Q_{\text{release}} = V_g C_o (-DH) Dt_{\text{cycle}}$$

$$Q_{\text{exhaust}} = V_g r_g C_p (T_{\text{exhaust}} - T_o) Dt_{\text{cycle}}$$

$Q$  is the quantity over the cycle,  $T_o$  is the inlet temperature,  $V_g$  is volumetric flow rate in ml/min,  $r_g$  is gas density in g/ml,  $C_o$  is concentration of the gas in mol/ml,  $C_p$  is gas capacity in J/g °C, and  $(-DH)$  is heat of reaction in J/mol. Using Equation 1, the heat loss for 10 % CO is 283.95 J/min or 64.82 % of heat released from the reaction.

The steady state experiments by Dhalewadikar (1984) for catalytic combustion of ethylene using supported platinum catalyst in a non-adiabatic fixed bed reactor also support the finding of this study. He shows that when the combustant concentration in the feed mixture is increased, the maximum temperature also increases. However, the ratio of the maximum temperature rise to the adiabatic temperature rise in his study also suggest a large heat loss in both the radial and axial directions. This researcher also found that the higher the combustant concentration, the greater is the heat loss from the system. By increasing the concentration of ethylene from 0.5 to 1 %, the ratio decrease from 18.09 to 15.11 %.

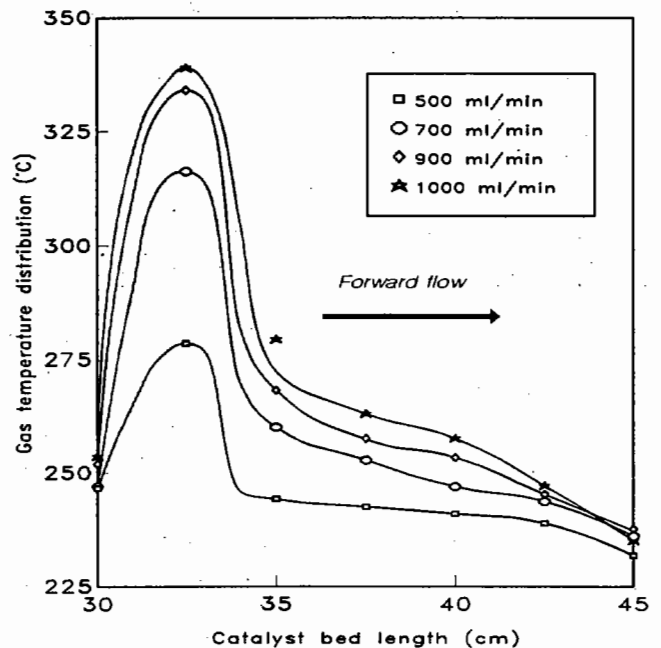


Figure 8. CO concentration effect on the temperature distribution

#### Effect of feed flow rate

To study the effect of feed flow rate, the flow was increased from 500 to 1000 ml/min while the inlet temperature (200 °C) and CO concentration (5%) were kept constant. This experiment was run for 15 cycles and the cycle time used was 30 min. The air coolant used was 1000 ml/min and the flow was countercurrent to the direction of the reactant mixture inside the reactor. The effect of feed flow rate on the maximum

measured temperature and heat removal is shown in Figures 9 and 10. From the figures, as expected, as the feed flow rate is increased, the maximum measured temperature and heat removal also increase. The figures show that by increasing the flow rate from 500 ml/min to 1000 ml/min, the maximum measured temperature increases by 60.6 °C for CO oxidation. The heat removal also increases by 6.34 J/min oxidation. Further variation of feed flow rate, between 500 and 1000 ml/min, demonstrate that the maximum measured temperature and heat removal are not linear in the flow rate. Figure 9 and 10 show the increase in both the maximum measured temperature and heat removal when the feed flow rate is increased. The temperature profiles for the variation of flow rate for both forward and reversed flows in the reaction zone at the end of 15th cycle are displayed in Figure 11.

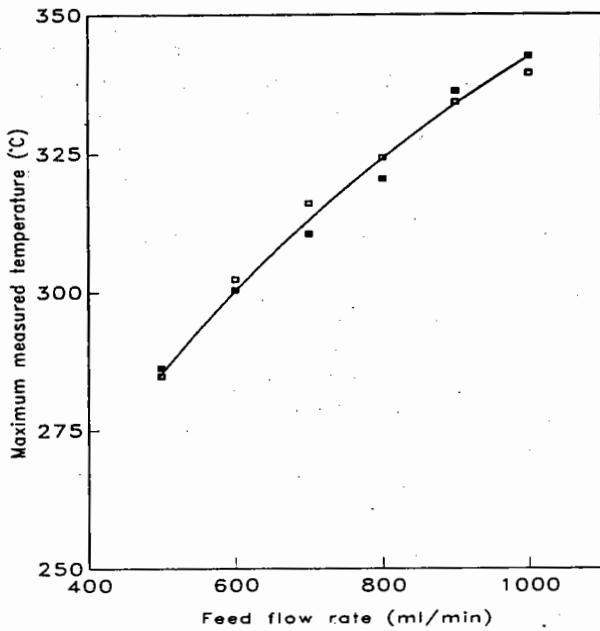


Figure 9. Effect of flow rate on maximum temperature

From Figures 11, it can be seen that the hot spot location is apparently unaffected by the flow rate in the flow rate range of 500 to 1000 ml/min which contrasts with the developing time-invariant thermal wave which usually moves upstream when the feed flow rate is increased as has been demonstrated by many researchers (Sharma and Hughes, 1979; and Puszynski and Hlavacek, 1980). The phenomenon in this study is expected since flow reversal always means ignition occurs at the beginning of the catalyst bed and does not depend on flow rate. Furthermore, the hot region is larger when the flow rate is higher. This is the result of a doubling of the heat release in the same amount of catalyst. For 100 % combustion with the same CO concentration, doubling the flow rate doubles the heat release. In addition, axial conduction and heat loss

would widen the profile. However, there is a possibility that the combustion is not complete near the bed entrance. If so, increasing the flow rate would shift the CO concentration profile downstream in the bed resulting in heat release further along the bed.

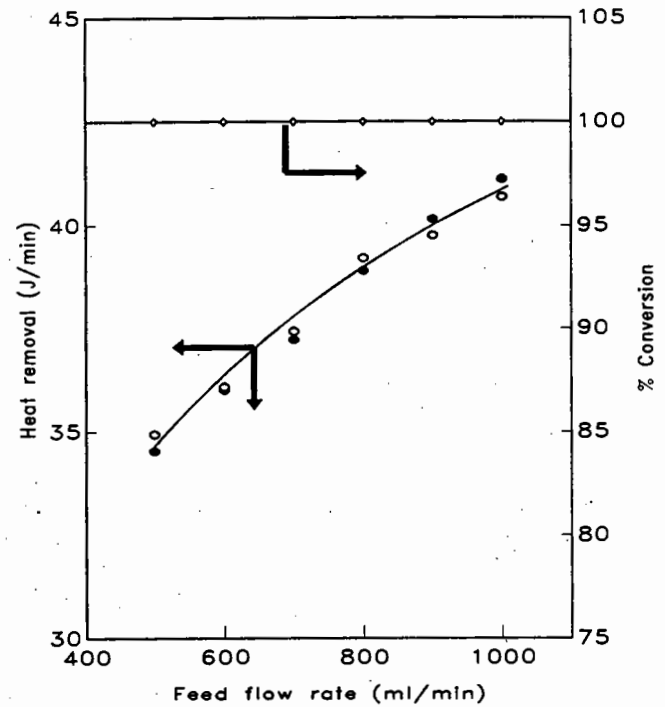


Figure 10. Effect of flow rate on heat removal and conversion

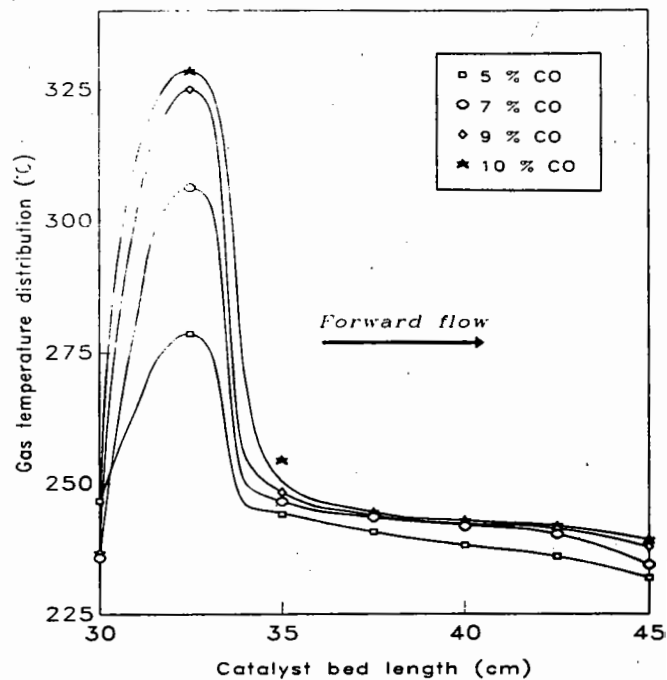


Figure 11. Effect of flow rate on the temperature distribution

From Figures 11; it can be seen that the hot spot location is apparently unaffected by the flow rate in the flow rate range of 500 to 1000 ml/min which contrasts with the developing time-invariant thermal wave which usually moves upstream when the feed flow rate is increased as has been demonstrated by many researchers (Sharma and Hughes, 1979; and Puszynski and Hlavacek, 1980). The phenomenon in this study is expected since flow reversal always means ignition occurs at the beginning of the catalyst bed and does not depend on flow rate. Furthermore, the hot region is larger when the flow rate is higher. This is the result of a doubling of the heat release in the same amount of catalyst. For 100 % combustion with the same CO concentration, doubling the flow rate doubles the heat release. In addition, axial conduction and heat loss would widen the profile. However, there is a possibility that the combustion is not complete near the bed entrance. If so, increasing the flow rate would shift the CO concentration profile downstream in the bed resulting in heat release further along the bed.

Figure 11 shows the expected phenomenon: that the maximum temperature increases when the feed flow rate is increased. The maximum temperature rise is related to the ratio of the heat released to the axial heat convection. This ratio is known as the Damkohler number. For this experiment,  $Da = Q(-r_A)d_p / C_p r_g V T = 4.903$ .  $Q$  is the heat released,  $(-r_A)$  is the reaction rate,  $d_p$  is the particle diameter,  $C_p$  is the gas heat capacity,  $r_g$  is the gas density,  $V$  is the volumetric flow rate and  $T_g$  is the gas temperature. According to Bathia (1991) and Gupta and Bathia (1991), in the mathematical analysis of the adiabatic flow reversal, the higher the Damkohler number, the higher the maximum bed temperature achieved. In addition, by doubling the flow rate, the heat release per unit catalyst volume about doubles and thus results in an increase of the maximum temperature. Therefore, the temperature profiles for 1000 ml/min flow rate should be the same as the temperature profiles for 10% CO. This study demonstrates that this is true, the temperature profiles for 1000 ml/min flow rate is the same as for 10% CO. Further variations of feed flow rate, between 500 and 1000 ml/min, demonstrate that the maximum measured temperature and heat removal are not linear in the flow rate.

The calculation of the ratio of maximum measured temperature rise to the adiabatic temperature rise and the degree of heat recovery shows that the ratio of maximum measured temperature rise to the adiabatic temperature rise increases as expected when the volumetric flow rates are increased. This is caused by the higher heat release for higher flow rate, but  $T_{ad}$  depends only on concentration, not on flow rate. Thus  $T_{ad}$  is the same for all  $Q$ . On the other hand, the percentage of heat removed decreases as the flow rate increases. This appears to be caused by the heat loss as discussed in the earlier section. Using Equation 1, the estimated heat loss for 1000 mL/min is 274.57 J/min or 62.68 % of heat released from the reaction. Results of mathematical modelling using adiabatic flow reversal

for combustion of low concentration of methane by Matros (1989) show that when the feed flow rate is increased, the maximum temperature increases while the degree of heat recovery decreases. Thus, these experimental results are in accordance with the model and offer model validation.

### Effect of initial temperature

To study the influence of inlet temperature, the bed was heated up to slightly above 250 °C. In this experiment, the CO concentration and the feed flow rate were kept at their minimum levels, all other conditions were as discussed for the base case. The result after 15th cycles is unexpected. By increasing the bed initial temperature from 200 to 250 °C, the maximum measured temperature increases by 37 °C. The heat removal also increases by 3.89 J/min. Intuitively (Matros, 1989), the effect of the initial temperature should not affect the maximum measured temperature after many cycles. This finding suggest that even 15 cycles provide insufficient time for the effect of initial bed temperature to disappear. Figure 12 shows that the maximum temperature in the bed decreases with increasing cycle number, which suggests that after a sufficient length of time, the temperature profiles will become independent of the initial bed temperature. The "wrong behaviour" that the measured maximum temperature rises first then declines may be caused by heat storage in the packing. The slow change is caused by need to dissipate the initial thermal energy through extra heat loss and no load on the coolant.

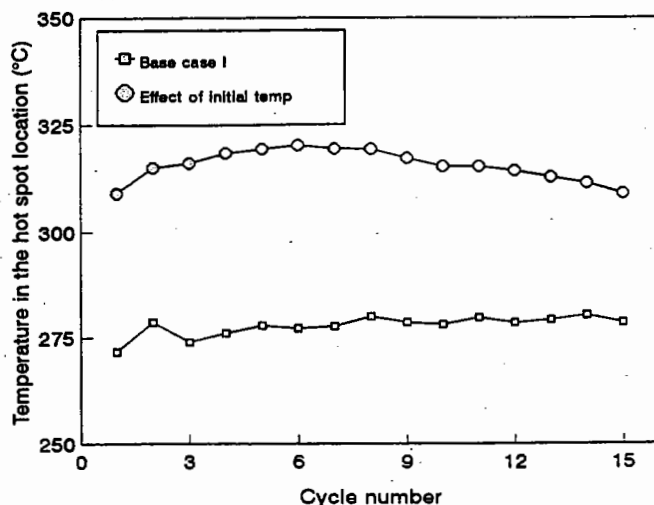


Figure 12. Effect of initial temperature on the progress of maximum temperature

### Effect of cycle time

At the otherwise minimum levels, the effect of cycle time at the end of 15th cycle on maximum measured temperature and heat removal have also been investigated. The result shows that cycle time has only a small effect on the maximum measured temperature and heat removal. If the cycle time are increased from 30 to

60 min, the maximum temperature and heat removal increase only 2.1 °C and 1.44 J/min for CO oxidation. This is expected, provided 60 minutes does not lead to blow out, because increasing the cycle period does not affect the heat release per unit catalyst volume. However, the small increase in the maximum measured temperature and heat removal is not supported by the flow reversal literature since two other behaviours were found for adiabatic processes regarding the dependency of the maximum temperature on the cycle time. Eigenberger and Nieken (1988), using simulation on a monolith catalyst for methane and propene combustion, found that by increasing the cycle period, the maximum temperature decreases. According to them, the longer the period, the more the heat accumulated tends to be transferred into the end sections of the reactor. In this study, this would be the recuperators ends. Matros (1989) studied the dependence of the maximum temperature on cycle duration for simulation of CO oxidation on oxide catalyst. He observed that the maximum temperature initially decreases with increasing cycle duration, passes through a minimum and then increases again once the cycle duration exceeds 25 mins. According to him, this behaviour occurs when the system achieves a configuration in which the rate of heat removals is a maximum.

#### CONCLUSIONS

A laboratory scale non-adiabatic-flow reversal reactor with integral heat exchange, using Pt/gamma-alumina catalyst was successfully operated on a dilute CO stream between 5 and 10 % CO. Integral heat exchange functional well, 15 to 25 % of the heat of reaction was removed from the system. The maximum bed temperature was experimentally found to be sensitive to changes in feed flow rate and reactant concentration. Experimentally, it was not sensitive to the length of a cycle.

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