

Carbon Monoxide Catalytic Oxidation Model

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Abstract

Carbon monoxide one of the major constituents of air pollution is released to the atmosphere mainly due to incomplete combustion of fuel. When released untreated to atmosphere it has harmful effects on human health. So in order to decrease its concentration in atmosphere, it has to be oxidized before its release. This can be achieved by use of catalytic converter. A mathematical model is formed in this paper to study the behavior of converter during the initial warm-up period. The model formed consists of mass and energy balance equations both for gas and solid phases. Results indicate the behavior of carbon monoxide conversion during the warm-up period along with variation of gas and solid temperatures in the converter assembly.

Keywords: Carbon monoxide, catalytic converter, Monolith, modeling.

1. Introduction

Hazardous chemicals escape to the environment by a number of natural and anthropogenic activities and may cause adverse effects on human health and the environment [Kampa and Castanas, 2008]. Carbon monoxide is one of the most common air pollutants present in the atmosphere today. Carbon monoxide is produced when fuel does not burn completely [Jie, 2011].

Pollution from automobile emissions has become over past few decades an issue of great concern. Production of carbon monoxide increases when cars are moving slowly hence levels of carbon monoxide in the atmosphere is higher in busy city streets than near highways. Levels may also increase in winter because cold engines emit much more carbon monoxide than hot engines [Hout, 1999]. Automotive emission control

comprises of technologies that are used to reduce the pollutants produced and released into the atmosphere by automobiles [Hascic, 2009].

Monolithic catalytic converter is a device that uses a catalyst to convert three harmful compounds in car exhaust into harmless compounds. As all the three major pollutants carbon monoxide, nitrogen oxides and hydrocarbons are simultaneously therefore, the catalyst used in the converter is called Three Way Catalytic Converter [Prasad and Singh, 2012].

Monolith comes from the Greek word “mono” means “single” and “lith” means “stone”. Monolith is large uniform block of a single building material of interconnected repeated cells A monolithic catalyst is most commonly made by applying the layer of catalytically active component (e.g. γ -alumina, SiO_2 , ZrO_2 , carbon, zeolites etc.) containing catalytically active components like Pt, Pd, Rh etc. are applied to the walls inside the walls of the basic insert monolith structure [Tomasic and Jovic, 2009]. Monolith have the advantages of low pressure drop, simple to construct, high mechanical strength, high geometric surface area [Chen and Ring, 2007].

In this paper, one dimensional model of carbon monoxide is to be considered, taking into account the mass and energy balance equations of gas and solid phase. The equations formed are a set of partial differential equations and these equations are coupled and solved using backward implicit scheme.

2. Mathematical Model

The oxidation reaction of carbon monoxide is considered:



The rate expression for catalytic reaction is given by [Yang et. al., 2008]:

$$-r_{CO} = k_0 \exp^{-E_a/RT} C_A^{-0.5} C_B^{0.74} \quad (2)$$

Where values of k_0 and E_a are $4.29 \times 10^4 \text{ gmol}^{0.77} \text{ cm}^{-1.31} \text{ s}^{-1}$ and $15.4 \text{ kcal/gmol}^{-1}$.

The final rate expression in terms of limiting reactant becomes:

$$-r_{CO} = k_0 e^{-E_a/RT} C_{A0}^{0.24} \left\{ \begin{array}{l} \left(\frac{C_A^4}{C_{A0}^4} \right) \left\{ 0.2734 M_0^{0.74} - \frac{1.233}{M_0^{0.26}} \frac{b}{a} - \frac{0.581}{M_0^{1.26}} \frac{b^2}{a^2} - \frac{0.673}{M_0^{2.26}} \frac{b^3}{a^3} - \frac{0.893}{M_0^{3.26}} \frac{b^4}{a^4} \right\} + \\ \left(\frac{C_A^3}{C_{A0}^3} \right) \left\{ -1.4061 M_0^{0.74} + \frac{3.221}{M_0^{0.26}} \frac{b}{a} + \frac{1.018}{M_0^{1.26}} \frac{b^2}{a^2} + \frac{0.9116}{M_0^{2.26}} \frac{b^3}{a^3} + \frac{0.974}{M_0^{3.26}} \frac{b^4}{a^4} \right\} + \\ \left(\frac{C_A^2}{C_{A0}^2} \right) \left\{ 2.953 M_0^{0.74} - \frac{4.6}{M_0^{0.26}} \frac{b}{a} - \frac{1.151}{M_0^{1.26}} \frac{b^2}{a^2} - \frac{0.815}{M_0^{2.26}} \frac{b^3}{a^3} - \frac{0.703}{M_0^{3.26}} \frac{b^4}{a^4} \right\} + \\ \left(\frac{C_A}{C_{A0}} \right) \left\{ -3.281 M_0^{0.74} + \frac{4.248}{M_0^{0.26}} \frac{b}{a} + \frac{0.7886}{M_0^{1.26}} \frac{b^2}{a^2} + \frac{0.431}{M_0^{2.26}} \frac{b^3}{a^3} + \frac{0.299}{M_0^{3.26}} \frac{b^4}{a^4} \right\} + \\ \left\{ 2.4609 M_0^{0.74} - \frac{1.82}{M_0^{0.26}} \frac{b}{a} - \frac{0.2367}{M_0^{1.26}} \frac{b^2}{a^2} - \frac{0.1}{M_0^{2.26}} \frac{b^3}{a^3} - \frac{0.056}{M_0^{3.26}} \frac{b^4}{a^4} \right\} \end{array} \right. \quad (4)$$

2.1 Assumptions

Some assumptions made during modelling include [Chauhan, 2011]:

- Monolithic is cylindrical with a circular cross section channel.
- Gas phase concentration, temperature and velocity are uniform across the monolithic cross section. Uniform properties are assumed for the flowing gas. Negligible axial diffusion of mass and heat transfer in gas phase
- Diffusion in washcoat is neglected, as washcoat is assumed to be very thin.
- The physical properties of monolith are constant and independent of monolithic temperature.
- Heat exchange between the substrate and the surroundings at both inlet and outlet faces of monolithic are neglected as there is lagging present at the inlet and outlet faces of the monolith.
- Heat exchange between the walls by radiation is neglected. Monolith is assumed to be adiabatic and heat transfer between the gas and the solid phase is by convection.

2.2 Modelling Equations

Mass balance in gas phase:

$$\frac{\partial C_A}{\partial t} = v \frac{\partial C_A}{\partial x} + a(-r_{CO}) \quad (5)$$

Energy Balance in Gas Phase:

$$-\rho_g C_{pg} v \left(\frac{\partial T_g}{\partial x} \right) - hS (T_g - T_s) = \rho_g C_{pg} \left(\frac{\partial T_g}{\partial t} \right) \quad (6)$$

Energy Balance in Solid Phase:

$$\rho_s C_{ps} \frac{\partial T_s}{\partial t} = \lambda_s \frac{\partial^2 T_s}{\partial x^2} + hS(T_g - T_s) + a(-\Delta H) (-r_{CO}) \quad (7)$$

Initial and Boundary Conditions

$$C_A(0,t) = C_A^0, T_g(0,t) = T_g^0, T_s(x,0) = T_s^0 \quad (8)$$

$$\text{At } x=0, \frac{\partial T_s}{\partial x} = 0 \text{ and at } x=L, \frac{\partial T_s}{\partial x} = 0, \frac{\partial C_A}{\partial x} = 0, \frac{\partial T_g}{\partial x} = 0 \quad (9)$$

All these above mentioned coupled equations are solved in dimensionless form and further convert into discretized form to form the final results.

3. Results and Discussions

Exhaust containing carbon monoxide enters the cold converter (initially at 25°C) and starts heating the converter. Once the operating temperature is achieved, the catalytic reaction starts. The inlet dimensionless concentration of carbon monoxide is taken as of 1.0000.

At a particular time as gas moves along the axial length, it comes in contact with more catalytic surface area thereby increasing its chance to react and hence its concentration decreases along the exit of converter length. Initially there is hardly any change in concentration of carbon monoxide as the converter has not attained the

operating temperature. At dimensionless time 25.80, the concentrations at axial distances 0.00, 0.20, 0.50 and 1.00 are 1.0000, 0.9538, 0.8884 and 0.7913 as shown in Figure 1. As dimensionless time increases to 28.90, the concentrations at same axial distance are 1.0000, 0.8132, 0.5435 and 0.0918 respectively.

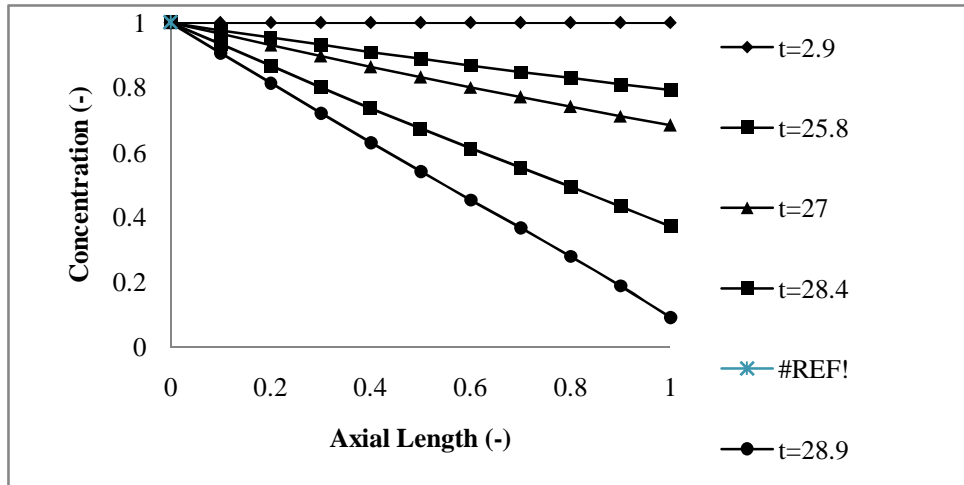


Figure 1: The effect of concentration variation along the converter length with time.

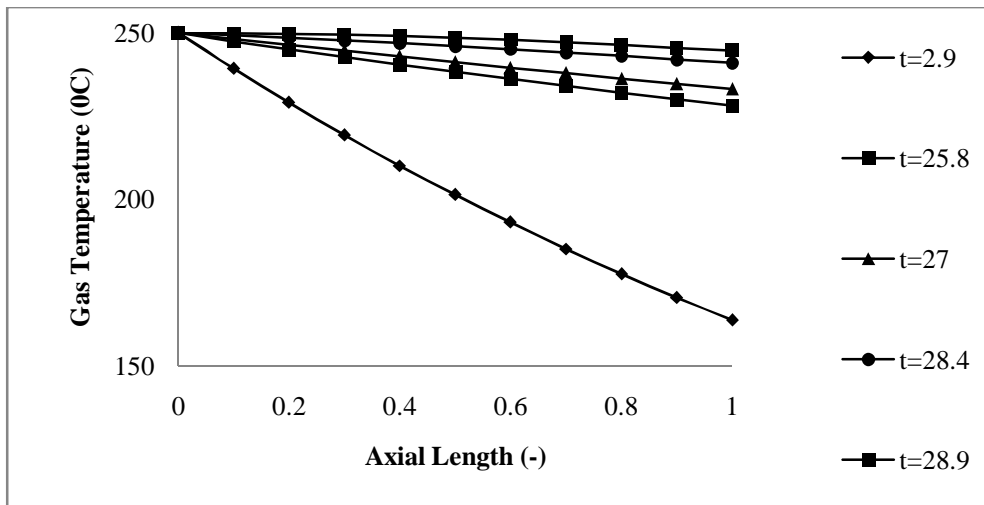


Figure 2: Gas temperature variation along the converter length with respect to time.

Gas temperature is more at the entrance and less near exit as shown in Figure 2. This is due to the loss of heat from gas as it flows along the length of the reactor. As dimensionless time increases, the gas temperature increases along the axial direction. At dimensionless time 2.90, the gas temperatures at axial distances 0.00, 0.20, 0.50 and 1.00 are 250.00°C, 229.05°C, 201.45°C and 163.98°C respectively. At dimensionless time 28.90, the gas temperatures at above stated axial distances are 250.00°C, 249.75°C, 248.61°C and 244.73°C respectively.

As dimensionless time increases, solid temperature increases due to heat supplied by the gas and later on due to onset of catalytic reaction. At dimensionless time 2.90, the solid temperature at axial distances 0.00, 0.20, 0.50 and 1.00 are 55.63^oC, 54.66^oC, 51.41^oC and 47.65^oC as shown in Figure 3. As dimensionless time increases to 28.90, the solid temperatures at same axial distances are 250.96^oC, 248.45^oC, 242.82^oC and 230.80^oC respectively.

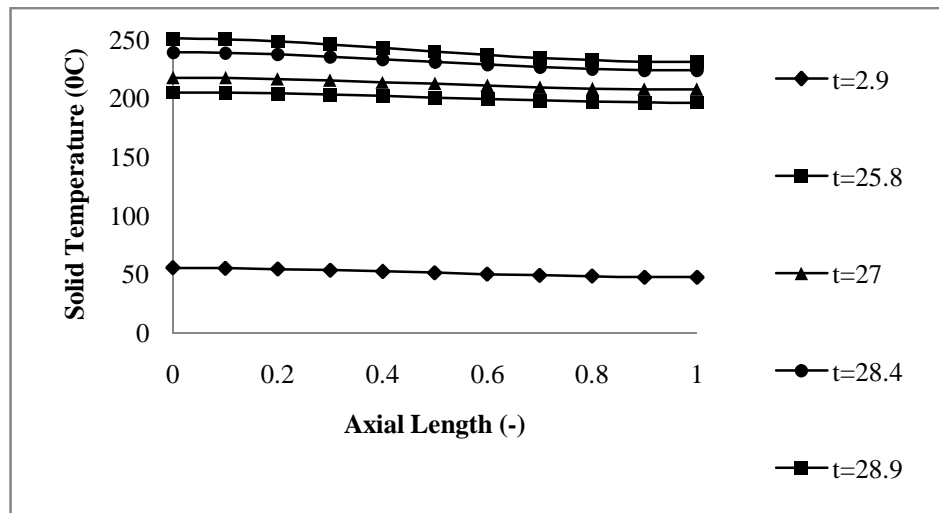


Figure 3: Solid temperature variation along the converter length with respect to time.

4. Conclusion

Development of a one-dimensional model for simulating the conversion of a oxidation of carbon monoxide is carried out. It is seen that appreciable amount of carbon monoxide emissions are released to the atmosphere during the warm up period. For carbon monoxide oxidation, it was observed that the concentration of the gas decreases as it moves along the length of the converter as it comes in contact with more catalyst surface area. It was observed that for carbon monoxide oxidation carried out in the converter channel, initially there is hardly any conversion of the polluting gas till the catalysts reach their operating temperatures This is due to the fact that initially when the converter is started, the solid temperature being very low; it does not favor the start of the catalytic reactions. As the gas temperature increases, the concentration decreases. As the solid temperature increases, the conversion of carbon monoxide increases.

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