

## **THREE WAY CATALYTIC SIMULATION OF ENGINE-OUT EXHAUST EMISSION**

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### **ABSTRACT**

*Presently Three Way Catalytic Converters (3WCC) are important after treatment devices mounted in the exhaust system of the engines to reduce harmful engine-out emissions. The experimental prediction of catalytic converter performance is time consuming and requires a lot of experimentation and large experimental set-up. In this paper, numerical simulation has been carried out for prediction of emission reduction. CHEMKIN 4.1 software is a powerful tool for solving complex chemical kinetics problems. Catalytic conversions of NO<sub>x</sub>, CO, and unburned hydrocarbons are numerically modeled by the Pt/Rh catalyst surface mechanisms in CHEMKIN 4.1. In the present work, exhaust emissions data of 2500 cc, spark ignition dedicated natural gas engine has been used to set dynamic inlet conditions for simulation. The simulation results show the conversion of unburned hydrocarbons (C<sub>3</sub>H<sub>6</sub>) and CO is, almost complete at the temperature of 709~710 K while 80% NO<sub>x</sub> is converted whereas it remains constant for higher temperature. These pollutants arrive at optimum conversion in about 93 sec.*

**Keywords:** *Engine-out emissions, Catalysts, Conversion Efficiency, 3WCC*

### **1) INTRODUCTION**

The numbers of automobile have increased many folds (about ten times) in the last fifty years which resulted in an enormous increase in urban air pollution level especially in developing countries. The pollutant emissions from automobile have been implicated in formation of green house gases, respiratory illness, and the depletion of the earth's ozone layer. The three pollutants of concern emitted from the spark ignition engines are unburned HC, CO and NO<sub>x</sub> (currently regulated).

Unburned HC and CO are generated due to inefficient combustion (when combustion efficiency less than 100%) while the thermal NO<sub>x</sub> is formed

when nitrogen reacts with excess oxygen at higher temperature ( $>1800\text{K}$ ) in the combustion process and is the predominant form produced by Zeldovich Mechanism in combustion engines (Mansha et al., 2012).

Two methods are being used to reduce harmful engine out exhaust emissions. One is to improve the engine technology & composition of fuels and introduction of alternative fuels so that better combustion occurs and consequently fewer emissions are generated. The second method is post combustion after treatment of the engine out exhaust gases.

In conventional spark ignition engines exhaust gas temperatures may vary from  $300^{\circ}\text{C}$  to  $400^{\circ}\text{C}$  from idling to around  $900^{\circ}\text{C}$  at full load condition. However, the most common range of exhaust gas temperature (EGT) lies between  $400^{\circ}\text{C}$ ~ $600^{\circ}\text{C}$ . Oxidation of unburned HC in the exhaust requires lower temperature than to oxidize carbon monoxide (CO) at a specified residence time. Without using catalyst, temperatures in excess of  $600^{\circ}\text{C}$  & residence time greater than 50ms is required to oxidize HC and temperature in excess of  $700^{\circ}\text{C}$  is required to oxidize CO (Heywood J.B., 1988). But, temperature ranges between  $250^{\circ}\text{C}$  and  $300^{\circ}\text{C}$  are required to oxidize CO & HC in the presence of catalysts.

Catalytic Converters are important post combustion after treatment devices mounted in the exhaust system of engines to reduce engine out exhaust emissions and are classified as Two-Way or Three-Way. Two-Way Catalytic Converter works on two gases, CO and unburned HC while the  $\text{NO}_x$  is controlled though exhaust gas recirculation (EGR) and by retarding the ignition timing.

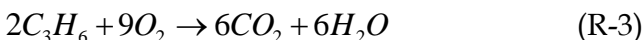
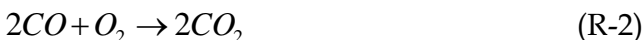
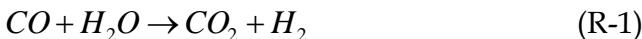
The Three-Way Catalytic Converter (3WCC) works on all the three gaseous pollutants of concern: CO, unburned HC &  $\text{NO}_x$ . 3WCC typically contain active catalytic materials (Pt/Rh or Pd/Rh) which promote oxidation of CO & unburned HC and reduction of  $\text{NO}_x$ . Pt (Platinum) and Pd (Palladium) are used as oxidation catalyst while Rh (Rhodium) is used as reducing catalyst for  $\text{NO}_x$  reduction. Other than precious metal catalysts, cheaper combination based on copper, chromium, nickel, manganese, etc have also been tried but could not achieve comparative efficiency. Research is still continued on the most promising copper based catalytic system (Kamble et al., 2008).

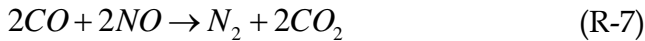
In three-way catalytic converter efficient diminution of CO, unburned HC and NO<sub>x</sub> are simultaneously carried out on the same catalyst bed. The maximum conversion efficiency can be achieved by operating the engine near stoichiometric condition though precise control of A/F mixture. If the mixture is too lean, NO<sub>x</sub> will not be destroyed and if the mixture is too rich CO & unburned HC will not be destroyed. The allowable A/F ratio variation window ( $\lambda$ -window) for efficient catalytic operation has been quoted by various researchers (Heywood, 1997); (Stone Richard, 1999) and (Ferguson, 2004) is as, Heywood, 0.1 ( $\pm 0.07\Phi$ ), Richard Stone  $\pm 0.25$  ( $\pm 0.02\Phi$ ) and Ferguson,  $\pm 1\%$  of stoichiometric. The precise control of A/F ratio has been achieved by electronic fuel metering using O<sub>2</sub> (lambda) sensor to provide feedback by measuring the oxygen concentration in the exhaust.

Ferguson (Ferguson, 2004) quoted the conversion efficiency at fully warmed up catalytic conditions about 98% to 99% for CO and 95% for unburned HC depending on HC components. Heywood quoted that fully warm up 3-way catalytic converter reduces the emission of unburned HC, CO & NO<sub>x</sub> by a factor of 10 and Willard W. Pulkrabek quoted that a fully warmed up catalytic converter at 400°C or higher will eliminate 98-99% of CO, 95% of NO<sub>x</sub> and more than 95% of unburned HC from the engine out exhaust.

The activation of the catalytic conversion does not take at the cold condition of exhaust emissions. In such conditions, the decrease in NO<sub>x</sub>, CO and unburned HC is not observed. The rate of oxidation of CO and HC increases and decrease in NO<sub>x</sub> takes place with increase of exhaust gasses. 50% conversion efficiency is called as light off of catalyst.

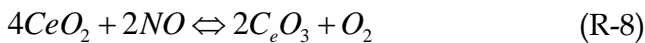
The chemical reactions, in warmed conditions, are as follows (Pattas et al, 1994):





The dynamics of a three-way catalytic converter also incorporate an oxygen storage process. The cerium oxide has the ability to store oxygen under weak operating conditions and release it during rich mixture condition (R-8). This is important for the operation of the three way catalyst, since the oxygen sensor can only determine whether a mixture is weak or rich, thus the engine control system seek to oscillate the air-fuel ratio around stoichiometric. By incorporating a second sensor downstream of catalyst and comparing the response with upstream sensor, the oxygen storage performance of the catalyst can be assessed (Riek, 1998). The oxygen can be stored by the following two mechanisms:

The chemical reaction is given below:



Equation (R-8) is the main oxygen storage system show into Three way conversion (Pattas, 1994), while the mechanism of (R-9) is a favorable outcome of catalytic material.

A catalytic converter besides its housing has three main components (a) Catalyst (b) Substrate or Support (c) Intermediate Coat or Wash Coat. The active catalysts (Pt, Pd, Rh) are supported on either a metal or ceramic substrate (Alumina  $Al_2O_3$ , Aluminum Oxide). The substrate is also covered with wash coat based on  $\gamma$ -  $Al_2O_3$  and containing stabilizers such as cerium oxide ( $CeO_2$ ) and barium oxide (BaO). The basic function of the substrate is to bring the active catalyst in to maximum contact with the exhaust gases.

Typical configurations of 3WCC include packed-bed and monolithic. The packed-bed catalytic converters used in 1970's passenger cars had a bed of spherical ceramics pellets made of  $\gamma$ -alumina. On the porous surface of pellets which provides a large surface area, the noble metal salts are impregnated. These converters give high pressure drop. The gas flow through the converter is a mix of axial and radial flow to provide large flow surface area to reduce back pressure.

The majority of catalytic converters used to day have a monolithic structure (ceramic honeycomb or metallic matrix). Monoliths are multi-channel structure through which the exhaust gas passes with a high volume flow rates. The channel walls are coated with a high surface porous material (wash coat) with finely dispersed noble metal catalytic particles (Platinum, Palladium or Rhodium) on it. High wash coat surface enables a high conversion despite low residence time.

## **2) NUMERICAL SIMULATION**

Numerical simulation is carried out using engine exhaust after treatment library of CHEMKIN 4.1 software. CHEMKIN 4.1 is a powerful tool for solving complex chemical kinetics problems. Catalytic conversions of  $\text{NO}_x$ , CO, and unburned hydrocarbons ( $\text{C}_3\text{H}_6$  -taken as represented to unburned hydrocarbon UHCs) are numerically modeled by the Pt/Rh, three-way catalyst surface mechanisms. A CHEMKIN 4.1 Honeycomb Monolith reactor combined with catalytic mechanism approximates a TWC. The system is designed in such a way with which the conversion of CO, HC and reduction in  $\text{NO}_x$  is possible through reaction on Pt/Rh. The chemistry of gas phase is ignored.

A 2.5L, 4 stroke 4-cylinders dedicated natural gas spark ignition engine having 13 compression ratio (CR-13) was tested on engine dynamometer for engine speeds of 1000, 1500, 2000, 2500 and 3000 rpm. The concentration of  $\text{NO}_x$ , UHCs, CO,  $\text{CO}_2$  &  $\text{O}_2$  (from engine exhaust) was measured using 5-gas exhaust analyzer. A thermocouple was fitted in the exhaust manifold just before the proposed location of the catalytic converter to measure the exhaust gas temperature. The engine out exhaust emission data is used for inlet conditions for an engine exhaust after treatment simulation. The software is is compiled and linked into the standard installation, so that it may work from the CHEMKIN 4.1 Interface without the help of a nay other compiler.

The reactor schematic model shows one gas inlet stirred reaction. (approximate 3WCC) is shown in Figure 1.

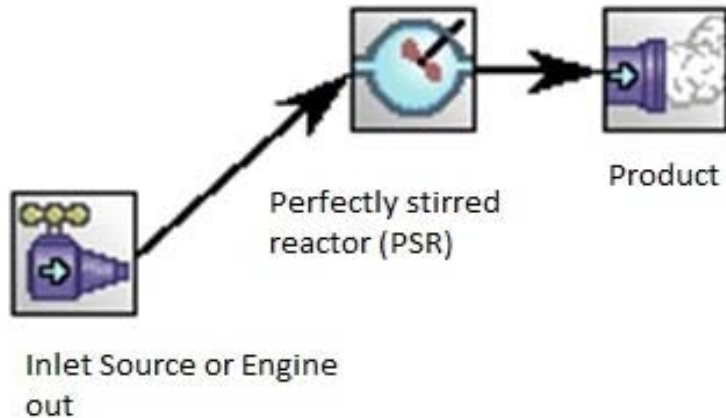


Figure 1: PSR after Treatment Diagram

A friendly user subroutine has been prepared which can be edited/modified as per requirements and availability of data. The contents of this file are given as input to the software and compositions, temperature, and exhaust flow rates are resulted.

## 2.1) Reactor Settings

In this typical problem, the Transient Solver is selected to solve Gas Energy equation. In the after treatment library, 1400cm<sup>3</sup> reactor volume has been considered which is the suitable amount of volume in the converter honeycomb or spongy medium for the entire length of the converter. In the same way, the convertor reactive area has been determined as 5.9×10<sup>7</sup> mm<sup>2</sup>. The area was determined based on valid geometry of the system. Due to specific volume and flow rate of gasses, the non-transient input time was considered. (CHEMKIN 4.1, 2004).

## 2.2) Surface Mechanism for Pt/Rh Reactions for 3WCC

There are two different kinds of adsorption sites:

Pt(S) = (S) and Rh (S1) = (S1), the total surface is made of 75% Pt and 25% Rh, i.e. the initial coverage must be 0.75 (S) and 0.25 (S1), the mechanism is valid only for this Pt/Rh ratio. In particular, the coverage dependent activation energies are scaled for that ratio (Chatterjee, 2004). Following are the surface mechanism for Pt/Rh reactions for three-way catalytic converter.

Reactions $k = A * T^{**b} * \exp(-Ea/RT)$	A (mol cm sec K)	b	Ea (cal/mol)
<b>1. C3H6 Oxidation on Pt</b>			
<b>A. Adsorption</b>			
O2 +Pt(S) +Pt(S) =>O(S) +Q(S)	7.000E02	0.0	0.0
C3H6 +Pt(S) +Pt(S) =>C3H6(S)	9.800E01	0.0	0.0
C3H6 +O(S) +Pt(S) =>C3H5(S) +OH(S)	5.000E02	0.0	0.0
H2 +Pt(S) +Pt(S) =>H(S) +H(S)	4.600E02	0.0	0.0
H2O +Pt(S) =>H2O(S)	7.500E01	0.0	0.0
CO2 +Pt(S) =>CO2(S)	5.000E03	0.0	0.0
CO +Pt(S) =>CO(S)	8.400E01	0.0	0.0
<b>B. Desorption</b>			
O(S) +Q(S) =>Pt(S) +Pt(S) +O2	3.700E+21	0.0	232.2
C3H6(S) =>C3H6 +Pt(S) +Pt(S)	1.000E+13	0.0	72.7
C3H5(S) +OH(S) =>C3H6 +O(S) +Pt(S)	3.700E+21	0.0	31.0
H(S) +H(S) =>H2 +Pt(S) +Pt(S)	3.700E+21	0.0	67.4
H2O(S) =>Pt(S) +H2O	1.000E+13	0.0	40.3
CO(S) =>CO +Pt(S)	1.000E+13	0.0	136.4
CO2(S) =>CO2 +Pt(S)	1.000E+13	0.0	27.1
<b>C. Surface reaction</b>			
C3H5(s) Oxidation [global reaction]			
C3H5(S) +5O(S) + 2Pt(S) =>5OH(S) +3C(S)	3.700E+21	0.0	95.0
<b>C3H6(s) Consumption</b>			
C3H6(S) =>CC2H5(S) +H(S)	1.000E+13	0.0	75.4
CC2H5(S) +H(S) =>C3H6(S)	3.700E+21	0.0	48.8
CC2H5(S) +Pt(S) =>C2H3(S) +CH2(S)	3.700E+21	0.0	108.2
C2H3(S) +CH2(S) =>CC2H5(S) +Pt(S)	3.700E+21	0.0	3.2
C2H3(S) +Pt(S) =>CH3(S) +C(S)	3.700E+21	0.0	46.0
CH3(S) +C(S) =>C2H3(S) +Pt(S)	3.700E+21	0.0	46.9
<b>CHx Consumption</b>			
CH3(S) +Pt(S) =>CH2(S) +H(S)	1.260E+22	0.0	70.4
CH2(S) +H(S) =>CH3(S) +Pt(S)	3.090E+22	0.0	0.0
CH2(S) +Pt(S) =>CH(S) +H(S)	7.000E+22	0.0	59.2
CH(S) +H(S) =>CH2(S) +Pt(S)	3.090E+22	0.0	0.0
CH(S) +Pt(S) =>C(S) +H(S)	3.090E+22	0.0	0.0
C(S) +H(S) =>CH(S) +Pt(S)	1.250E+22	0.0	138.0
<b>C2Hx Oxidation</b>			
C2H3(S) +O(S) =>Pt(S) +CH3CO(S)	3.700E+19	0.0	62.3
CH3CO(S) +Pt(S) =>C2H3(S) +O(S)	3.700E+21	0.0	196.7
CH3(S) +CO(S) =>Pt(S) +CH3CO(S)	3.700E+21	0.0	82.9
CH3CO(S) +Pt(S) =>CH3(S) +CO(S)	3.700E+21	0.0	0.0
<b>CHx Oxidation</b>			
CH3(S) +O(S) =>CH2(S) +OH(S)	3.700E+21	0.0	36.6

$\text{CH}_2(\text{S}) + \text{OH}(\text{S}) \Rightarrow \text{CH}_3(\text{S}) + \text{O}(\text{S})$	3.700E+21	0.0	25.1
$\text{CH}_2(\text{S}) + \text{O}(\text{S}) \Rightarrow \text{CH}(\text{S}) + \text{OH}(\text{S})$	3.700E+21	0.0	25.1
$\text{CH}(\text{S}) + \text{OH}(\text{S}) \Rightarrow \text{CH}_2(\text{S}) + \text{O}(\text{S})$	3.700E+21	0.0	25.2
$\text{CH}(\text{S}) + \text{O}(\text{S}) \Rightarrow \text{C}(\text{S}) + \text{OH}(\text{S})$	3.700E+21	0.0	25.1
$\text{C}(\text{S}) + \text{OH}(\text{S}) \Rightarrow \text{CH}(\text{S}) + \text{O}(\text{S})$	3.700E+21	0.0	224.8
<b>H, OH, H<sub>2</sub>O Surface Reactions</b>			
$\text{O}(\text{S}) + \text{H}(\text{S}) \Rightarrow \text{OH}(\text{S}) + \text{Pt}(\text{S})$	3.700E+21	0.0	11.5
$\text{OH}(\text{S}) + \text{Pt}(\text{S}) \Rightarrow \text{O}(\text{S}) + \text{H}(\text{S})$	5.770E+22	0.0	74.9
$\text{H}(\text{S}) + \text{OH}(\text{S}) \Rightarrow \text{H}_2\text{O}(\text{S}) + \text{Pt}(\text{S})$	3.700E+21	0.0	17.4
$\text{H}_2\text{O}(\text{S}) + \text{Pt}(\text{S}) \Rightarrow \text{H}(\text{S}) + \text{OH}(\text{S})$	3.660E+21	0.0	73.6
$\text{OH}(\text{S}) + \text{OH}(\text{S}) \Rightarrow \text{H}_2\text{O}(\text{S}) + \text{O}(\text{S})$	3.700E+21	0.0	48.2
$\text{H}_2\text{O}(\text{S}) + \text{O}(\text{S}) \Rightarrow \text{OH}(\text{S}) + \text{OH}(\text{S})$	2.350E+20	0.0	41.0
<b>CO Oxidation</b>			
$\text{CO}(\text{S}) + \text{O}(\text{S}) \Rightarrow \text{CO}_2(\text{S}) + \text{Pt}(\text{S})$	3.700E+20	0.0	108.0
$\text{CO}_2(\text{S}) + \text{Pt}(\text{S}) \Rightarrow \text{CO}(\text{S}) + \text{O}(\text{S})$	3.700E+21	0.0	165.1
$\text{C}(\text{S}) + \text{O}(\text{S}) \Rightarrow \text{CO}(\text{S}) + \text{Pt}(\text{S})$	3.700E+21	0.0	0.0
$\text{CO}(\text{S}) + \text{Pt}(\text{S}) \Rightarrow \text{C}(\text{S}) + \text{O}(\text{S})$	3.700E+21	0.0	218.5
<b>2. NO Reduction on Pt</b>			
<b>A. Adsorption</b>			
$\text{NO} + \text{Pt}(\text{S}) \Rightarrow \text{NO}(\text{S})$	8.500E01	0.0	0.0
<b>B. Desorption</b>			
$\text{NO}(\text{S}) \Rightarrow \text{NO} + \text{Pt}(\text{S})$	1.000E+16	0.0	140.0
$\text{N}(\text{S}) + \text{N}(\text{S}) \Rightarrow \text{N}_2 + \text{Pt}(\text{S}) + \text{Pt}(\text{S})$	3.700E+21	0.0	113.9
<b>C. Surface Reaction</b>			
$\text{NO}(\text{S}) + \text{Pt}(\text{S}) \Rightarrow \text{N}(\text{S}) + \text{O}(\text{S})$	5.000E+20	0.0	107.8
$\text{N}(\text{S}) + \text{O}(\text{S}) \Rightarrow \text{NO}(\text{S}) + \text{Pt}(\text{S})$	3.700E+21	0.0	128.1
<b>3. NO Reduction and CO Oxidation on Rhodium</b>			
<b>A. Adsorption</b>			
$\text{O}_2 + \text{Rh}(\text{S1}) + \text{Rh}(\text{S1}) \Rightarrow \text{O}(\text{S1}) + \text{O}(\text{S1})$	1.000E02	0.0	0.0
$\text{CO} + \text{Rh}(\text{S1}) \Rightarrow \text{CO}(\text{S1})$	5.000E01	0.0	0.0
$\text{NO} + \text{Rh}(\text{S1}) \Rightarrow \text{NO}(\text{S1})$	5.000E01	0.0	0.0
<b>B. Desorption</b>			
$\text{O}(\text{S1}) + \text{O}(\text{S1}) \Rightarrow \text{O}_2 + \text{Rh}(\text{S1}) + \text{Rh}(\text{S1})$	3.000E+21	0.0	293.3
$\text{CO}(\text{S1}) \Rightarrow \text{CO} + \text{Rh}(\text{S1})$	1.000E+14	0.0	132.3
$\text{NO}(\text{S1}) \Rightarrow \text{NO} + \text{Rh}(\text{S1})$	5.000E+13	0.0	108.9
$\text{N}(\text{S1}) + \text{N}(\text{S1}) \Rightarrow \text{N}_2 + \text{Rh}(\text{S1}) + \text{Rh}(\text{S1})$	1.110E+19	0.0	136.9
<b>C. NO/CO Surface Reaction</b>			
$\text{CO}(\text{S1}) + \text{O}(\text{S1}) \Rightarrow \text{CO}_2 + \text{Rh}(\text{S1}) + \text{Rh}(\text{S1})$	3.700E+20	0.0	59.9
$\text{NO}(\text{S1}) + \text{Rh}(\text{S1}) \Rightarrow \text{N}(\text{S1}) + \text{O}(\text{S1})$	2.220E+22	0.0	79.5
END			



### 3) RESULTS AND DISCUSSIONS

#### 3.1) Conversion Rate Vs Temperature

In Fig. 2, the conversion rate of CO is shown as function of temperature. The conversion of CO starts at 573K (300°C) and increases up to 98% at 427°C (700K).

In Fig. 3, the conversion of  $C_3H_6$  starts at 600K (327°C), but increases more slowly with temperature as compared to conversion rate of CO. At 700 K the conversion of  $C_3H_6$  is 70% as compared to CO conversion of 98%. The conversion of  $C_3H_6$  is complete at 710K.

In Fig. 4 and 5, the conversion of NO starts at higher temperature 704K (431°C) as compared with CO and  $C_3H_6$ . At 709 K the conversion of NO is 80% complete and remain at the same value (80%) at higher temperatures.

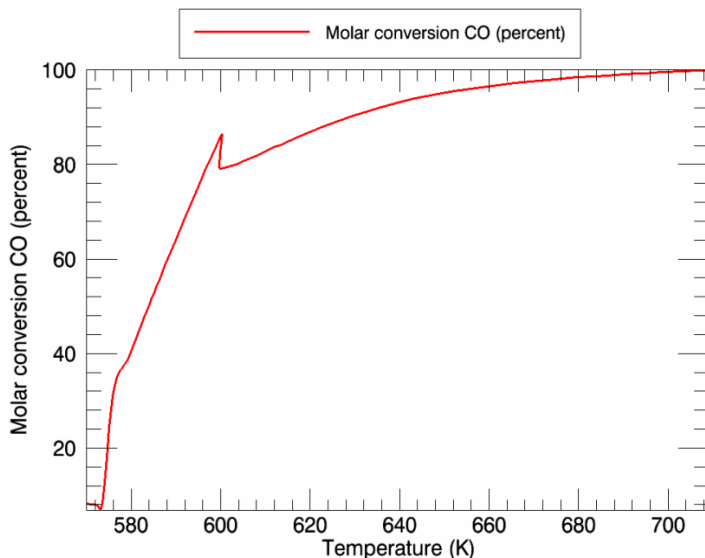


Figure 2: Molar Conversion of CO with Temperature

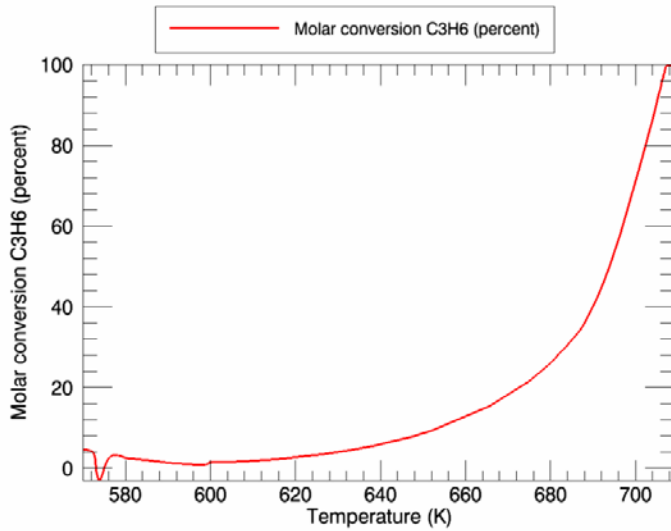


Figure 3: Molar Conversion of C3H6 with Temperature

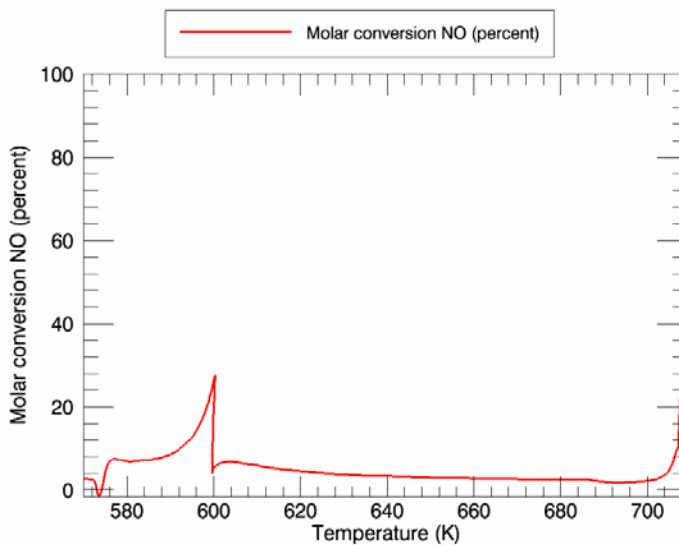


Figure 4: Molar Conversion of NO with Temperature

### 3.2) Conversion Rate Vs Time

Figure-5 shows molar conversions of C<sub>3</sub>H<sub>6</sub>, CO & NO with respect to time. Note that “molar conversion” variables must be selected on the Select Post-Processing Variables panel in order for molar conversion variables to be available for plotting.

Figure -5 clearly shows that  $C_3H_6$  and CO are converted more effectively than NO under these conditions and the three pollutants take about 93 sec to arrive at optimum conversion.

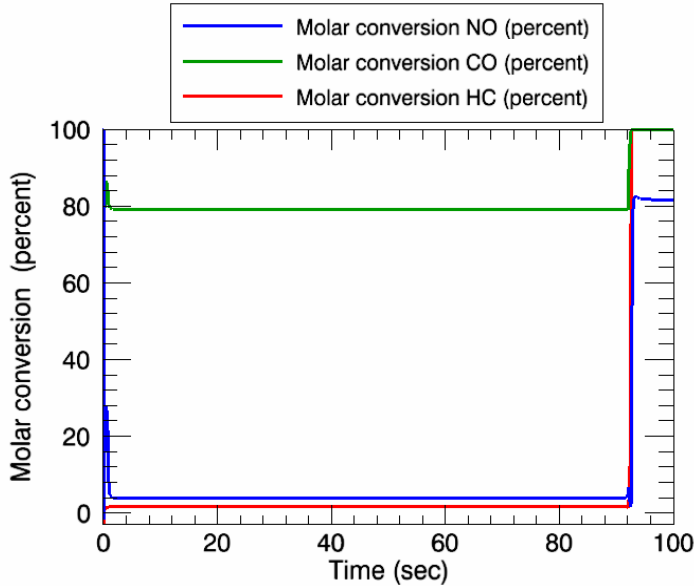


Figure 5: Molar Conversions of Emissions with Time

#### 4) CONCLUSIONS & FUTURE RECOMMENDATION:

Catalytic conversions of  $NO_x$ , CO, and UHCs ( $C_3H_6$ ) are numerically modeled by the Pt/Rh three way catalyst surface mechanisms through engine exhaust after treatment library in CHEMKIN 4.1 software. A detailed reaction mechanism based on chemistry module DETCHEM has been used for the conversion of CO,  $C_3H_6$ , and NO to simulate the exhaust gas treatment in a platinum/rhodium coated channel of a typical 3-way catalytic converter.

The computational tool has used to predict conversion rates at nearly stoichiometric conditions as function of temperature and time. Simulation results show that at 700 K the conversion of  $C_3H_6$  is 70% as compared to CO conversion of 98%. The conversion of  $C_3H_6$  is complete at 710K. The conversion of NO starts at higher temperature 704K (431°C) as compared with CO and  $C_3H_6$ . At 709 K the conversion of NO is 80% complete and remains at the same value (80%) at higher temperatures. In summary, it is concluded that having incorporated 3WCC in the exhaust

system of the engine, the emitting exhaust pollutants hydrocarbons ( $C_3H_6$ ) and CO are nearly converted while NO is 80% converted and stays at the same value at further higher temperature. Further the three pollutants take about 93 sec to arrive at this optimum conversion.

It is recommended that 3WCC be mounted in the exhaust pipe of this very engine for further reduction of the engine out exhaust emission to minimum level.

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