UNIVERSITI PUTRA MALAYSIA

NUMERICAL SIMULATION OF NITROUS OXIDE AND CARBON MONOXIDE ABATEMENT IN THE CATALYTIC CONVERTER OF A COMPRESSED NATURAL GAS ENGINE

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By

CHEONG YOON KWAN

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science

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Dedication

To my wife whose support and understanding helped make this possible; my sons for their cooperation and understanding especially on the lost weekends.

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

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September 2007

Chairman : Nor Mariah Bte Adam, PhD

Faculty : Engineering

Air pollution in Malaysia is mainly caused by emission from motor vehicles according to the Department of Environment (DOE). The Malaysia Government through DOE has been regulating vehicles emissions with more stringent regulations on nitrogen oxides and carbon monoxide emissions gazetted since 1996. Catalytic converters are one of the effective devices to reduce air pollution by motor vehicles by transforming nitrogen oxides and carbon monoxide in the exhaust to relatively harmless nitrogen and carbon dioxide respectively. A catalytic converter is easy to fit into any exhaust system, not bulky and does not require much maintenance. However, due to the presence of precious group metals such as platinum, rhodium and palladium as catalysts, the cost of the catalytic converter is relatively high. Furthermore, the catalysts' activation and deactivation levels is highly dependent on temperature, hence the design of catalytic converter in the vehicle exhaust system is not easy.

The objective of this study is to carry out simulation via CFD code FLUENT 6.0, on catalytic converter design and efficiency in a cold start natural gas engine for nitrogen monoxide and carbon monoxide emission control.

CFD code FLUENT 6.0 was used for prediction of catalytic converter light-off temperature and efficiency. Cold start and light-off temperatures are the acceptable worst scenario for compressed natural gas (CNG) engine pollutants abatement in order to achieve low emission vehicle. The simulation result was then verified via experimental data published in the literature. Another CFD modelling module was conducted to predict the exhaust gas temperature at 10cm, 30cm, 50cm, 80cm and 110cm from the engine outlet to determine the best position of catalytic converter in the exhaust system. A third CFD modelling module was done to simulate the surface reactions on a single channel of a catalytic converter. This is the contribution to the knowledge in the pollutants abatement in catalytic converter. The simulation result was then verified via experimental data published in the literature.

The simulation of catalytic converter light-off temperature for NO and CO were proved to be satisfactory when compared to presented experimental result. Simulated NO conversion efficiency was in agreement with presented experimental result. However, CO conversion simulation result was not well predicted compared with presented experimental result. This is because FLUENT 6.0 surface reaction does not take surface coverage into account. Simulation of exhaust gas temperature showed that it is not advisable to place the catalytic converter below 25cm or above 80cm from the engine outlet. It is found that CFD FLUENT 6.0 can be used to simulate surface reaction on a single channel by adjusting the Arrhenius constants by a factor of 10^{-15} for CO reaction and factor of 10^{-16} for NO reaction.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

SIMULASI BERANGKA RAWATAN NITROGEN OKSIDA DAN KARBON MONOSIDA DALAM PENUKAR BERMANGKIN UNTUK ENJIN GAS ASLI TERMAMPAT

Oleh

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Punca utama pencemaran udara di Malaysia adalah daripada kenderaan mengikut data Jabatan Alam Sekitar (JAS). Kerajaan Malaysia, melalui JAS telah mula memantau perlepasan nitrogen oksida and karbon monoksida daripada kenderaan dengan lebih ketat sejak diwartakan pada 1996. Penukar bermangkin merupakan salah satu alat yang berkesan untuk mengurangkan pencemaran udara yang disebabkan oleh kenderaan bermotor dengan menukar nitrogen dioksida dan karbon monoksida masing-masing kepada nitrogen dan karbon dioksida yang kurang berbahaya. Penukar bermangkin mudah dipasang di dalam sistem ekzos, bersaiz kecil dan tidak memerlukan banyak penyelenggaraan. Walau bagaimanapun, kos penukar bermangkin agak tinggi disebabkan kehadiran unsur platinum, rodium dan palladium. Tahap keaktifan dan nyah-aktif pemangkin bergantung kepada suhu dan ini menyebabkan rekabentuk penukar bermangkin dalam sistem ekzos kenderaan menjadi rumit.

Objektif kajian ini adalah untuk membuat simulasi menggunakan kod CFD FLUENT 6.0 terhadap rekabentuk dan kecekapan penukar bermangkin dalam menghidupkan

enjin gas asli dalam keadaan sejuk untuk mengawal perlepasan nitrogen monoksida dan karbon monoksida.

Kod CFD FLUENT 6.0 telah digunakan untuk meramal suhu mula tindakbalas dan kecekapan penukar bermangkin. Menghidupkan engine dalam keadaan sejuk dan meramal suhu mula tindakbalas adalah keadaan terburuk yang diperakui dalam pengawalan perlepasan pencemar oleh enjin dihidup dalam keadaan sejuk untuk mencapai tahap kenderaan perlepasan pencemar rendah (Heck et al., 2002). Keputusan simulasi yang terhasil membuktikan kesahihannya dengan keputusan eksperimen dari literatur. Satu modul simulasi CFD yang lain telah dibuat untuk meramal suhu gas ekzos pada kedudukan 10sm, 30sm, 50sm, 80sm dan 110sm dari enjin untuk menentukan kedudukan terbaik untuk memasang penukar bermangkin dalam sistem ekzos. Modul simulasi CFD ketiga dibuat untuk simulasi tindakbalas kimia permukaan pada satu salur dalam penukar bermangkin. Keputusan simulasi yang terhasil membuktikan kesahihannya dengan keputusan eksperimen dari literatur. Simulasi suhu mula tindakbalas untuk NO dan CO dalam penukar bermangkin didapati menepati keputusan eksperimen yang telah dilakukan. Hasil simulasi kecekapan penukaran NO juga didapati menyamai keputusan eksperimen. Walau bagaimanapun, hasil simulasi kecekapan penukaran CO tidak menyamai keputusan eksperimen dengan baik. Ini adalah disebabkan oleh FLUENT 6.0 yang tidak mengambil kira liputan permukaan tindakbalas. Hasil simulasi suhu gas eksos mendapati penukar bermangkin tidak digalakkan dipasang kurang daripada 25sm dari enjin dan juga melebihi 80sm dari enjin. Hasil penyelidikan ini juga mendapati CFD FLUENT 6.0 boleh digunakan untuk simulasi tindakbalas permukaan pada satu salur dengan melaras pekali Arrhenius dengan faktor 10^{-15} untuk tindakbalas CO dan 10^{-16} untuk tindakbalas NO.

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I certify that the Examination Committee has met on 4 September 2007 to conduct the final examination of Cheong Yoon Kwan on his Master of Science thesis entitled "Numerical Simulation of Nitrous Oxide and Carbon Monoxide Abatement in the Catalytic Converter of a Compressed Natural Gas Engine" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the student be awarded the degree of Master of Science.

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

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Date: 21 February 2008

DECLARATION

I hereby declare that the thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at UPM or other institutions.

CHEONG YOON KWAN

Date: 3 December 2007

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LIST OF ABBREVIATIONS

l	air fuel ratio
v	velocity (cm/s)
C	molar concentration (g.mol/cm^3)
x	length of catalytic converter (cm)
r	Rate of reaction (g.mol/s.cm^3)
k`	apparent rate constant
C_o	Outlet molar concentration (g.mol/cm^3)
C_i	inlet molar concentration (g.mol/cm^3)
t	Residence time (s)
K_g	mass transfer coefficient (cm/s)
a_v	geometric surface area per unit volume (cm^2/cm^3)
N_{Sh}	Sherwood number
N_{Sc}	Schmidt number
N_{Re}	channel Reynolds number
D	diffusivity of pollutant in air (cm^2/s)
L	honeycomb length (cm)
W	mass flowrate to honeycomb catalyst (g/s)
M	mass of gas flowing into monolith (g)
A	frontal area of honeycomb (cm^2)
d_{ch}	hydraulic diameter of honeycomb channel (cm)
r_g	gas density (g/cm^3)
m	gas viscosity (g/s.cm)
e	void fraction of honeycomb, dimensionless
H	gas heat transfer coefficient
T_s	monolith temperature(K)

T_g	gas temperature(K)
h_g	gas enthalpy(J/g)
C_{pg}	gas specific heat capacity(J/g.K)
C_{gi}	mole fraction of species i in the gas
C_{si}	mole fraction of species i on the surface of the monolith
K_{gi}	mass transfer coefficient of species i
M_i	molecular mass of species i
a_c	catalyst area per unit reactor volume(cm^2/cm^3)
R_i	chemical reaction rate of species i(g.mol/s.cm^3)
α	monolith porosity
r_s	monolith density(g/cm^3)
c_{ps}	monolith specific heat capacity(J/g.K)
k_s	monolith thermal conductivity(J/cm.K)
k_{eff}	effective thermal conductivity of the composite of exhaust gas and substrate
s_1	heat transfer between the monolith and the gas
s_2	heat released from chemical reactions
DH_i	the reaction heat of species i
k_g	number of elementary gas phase reactions
k_{fk}	forward rate coefficient
$[X_j]$	concentration of species j
A_r	pre-exponential factor
$v'_{i,r}$	stoichiometric coefficient for reactant *i* in reaction *r*
$v''_{i,r}$	stoichiometric coefficient for product *i* in reaction *r*

$D_{i,m}$	diffusion coefficient for species *i*
k	thermal conductivity
$\hat{R}_{i,r}$	Arrhenius molar rate of creation or destruction of species *i* in reaction *r*
$k_{f,r}$	forward rate constant for reaction *r*
$k_{b,r}$	backward rate constant for reaction *r*
$n'_{j,r}$	forward rate exponent for each reactant and product species *j* in reaction *r*
$n''_{j,r}$	backward rate exponent for each reactant and product species *j* in reaction *r*
Γ	net effect of third bodies on the reaction rate
$g_{j,r}$	third-body efficiency of the *j*th species in the *r*th reaction
K_r	equilibrium constant for the r^{th} reaction
p_{atm}	atmospheric pressure
S_i^0	standard-state entropy
h_i^0	standard-state enthalpy
N	total number of chemical species
$\vec{n}$	unit vector normal to the surface
$\dot{m}_{dep}$	total mass deposition rate
$Y_{i,wall}$	mass fraction of species *i* at the wall
br	temperature coefficient
E_r	activation energy
R	gas constant
S_i	rate of creation by addition from the dispersed phase
J_i	diffusion flux of species i
Y_i	local mass fraction of species i

CHAPTER 1

INTRODUCTION

1.1 Air Pollution

Air pollution is contamination of the atmosphere by gaseous, liquid, or solid wastes or by-products that can endanger human health as well as the health and welfare of plants and animals, or can attack materials, reduce visibility, or produce undesirable odours. Before 1980s, little attention was paid to air pollution in Malaysia. The agriculture sector and mining industries were still dominant then and manufacturing sector was still in its infant stage. Automobile population was much smaller before 1980s compared to now.

In haze-free time, air pollution in Malaysia is contributed from three major sources; i.e. vehicles, stationary sources (i.e. power stations, industries) and open burning sources. According to Rafia et al. (2002) from 1996 to 2001, the main air pollution source was from vehicles, contributing 70-75% of the total air pollution. Emissions from stationary sources contributed 20-25% of the air pollution. Open burning including forest fires contributed 3-5%. However, vehicles are contributing more pollution ever since. According to the Department of Environment Malaysia (DOE) in 2004, the percentages of the air emission load for carbon monoxides (CO), nitrous oxides (NOx), sulphide oxide (SO_2) and particulate matter (PM_{10}) were vehicles

88.3%; stationary sources 9.6% and others 2.1% as shown in Figure 1.1. Detailed emission of air pollutants in Malaysia is shown in Table 1.1.

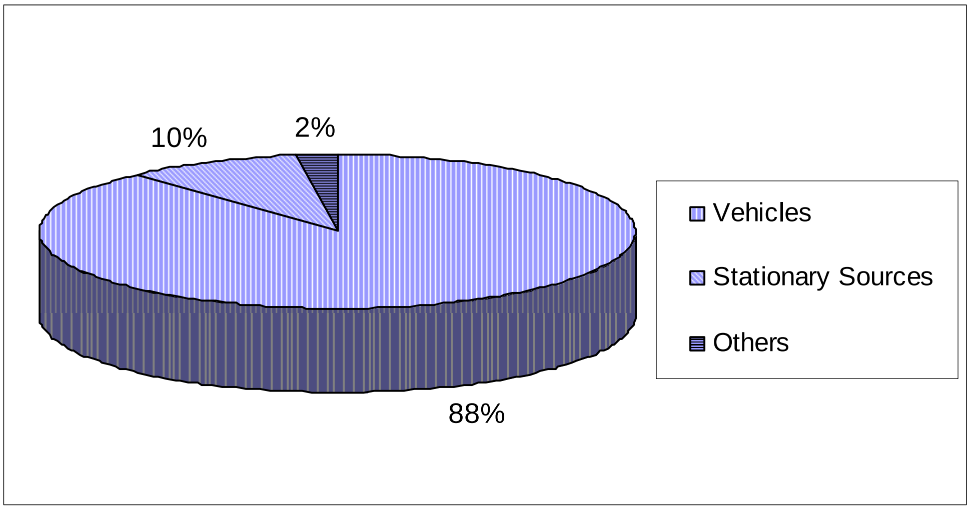

Figure 1.1: Sources of Air Pollution in Malaysia (DOE, 2005)

Table 1.1: Emission of Air Pollutants in Malaysia in 2004 (DOE, 2005)

Source	CO (ton)	NOx (ton)	SO_2 (ton)	Particulate Matter, PM_{10} (ton)	Total (ton) / (%)
Vehicles	1,241,249	182,281	136,156	20,151	1,579,837 (88.3%)
Stationary sources	33,274	116,585	11,624	9,418	170,901 (9.6%)
Others	5,640	9,537	22,016	409	37,602 (2.1%)
Total	1,280,163	308,403	169,796	29,978	1,788,340 (100%)

Vehicles include private cars, taxis, commercial vehicles and motorcycles. There were 9.1 million vehicles in-use in Malaysia at the end of 2004 compared to 7.7 million vehicles in 1996; an increase of 18 % (DOE, 2005). Figure 1.2 shows the number of in-use vehicles in 2004.

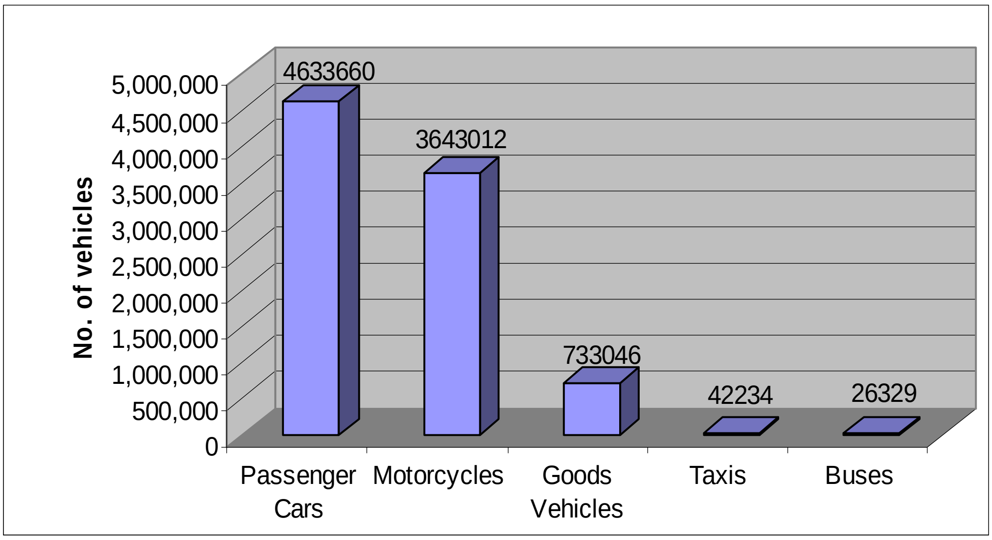

Figure 1.2: Number and Type of Vehicles in Malaysia in 2004 (DOE, 2005)

Geographically, Wilayah Persekutuan Kuala Lumpur has the highest number of vehicles (2.9 million) while Johor is second followed by Selangor, Pulau Pinang and Perak. Among the vehicles in Malaysia, private cars are the major contributor to air pollution. Motor vehicles contributed 98% of the total CO; 59% of total NOx; 31% of total particulate matter as well as 7% of the total SO_2 (DOE, 2005).

CO is known to cause poisoning to human by combining with hemoglobin in the blood in the lung and reducing oxygen level in the blood. NOx undergo photochemical reaction in the sunlight leading to generation of ozone and smog. (Air Quality Technical Report 43, 2004).

1.2 Regulation on Air Pollution

In order to reduce the impact of vehicles' emission on air quality, the Malaysian government has phased out the sales of leaded petrol. The sale of leaded petrol has been reduced since 1993 and was completely phased out by end of 1998. Reduced leaded gasoline sales are believed to be responsible for a steady decrease in lead levels throughout the western side of Peninsular Malaysia (Rafia et al., 2002). The Environmental Quality Act 1974 (EQA) has been amended to add three regulations designed to reduce emissions from vehicular sources. They are the Environmental Quality (Control of Emissions From Diesel Engines) Regulation 1996, the Environmental Quality (Control of Emissions From Petrol Engines) Regulation 1996 and recently the Environmental Quality (Control of Emissions From Motorcycles) Regulation 2003.

Environmental Quality (Control of Emissions from Diesel Engines) Regulation 1996 is applicable to all new diesel engine vehicles registered after August 1996 and to existing diesel engine vehicle with new engine replacing an existing engine. The regulations cover among others, the emission standard for visible pollutants, the emission standard for gaseous pollutants and smoke emission control.

For motorcycles, their emissions are governed under Environmental Quality (Control of Emission from Motorcycles) Regulations 2003. The regulations cover among others, the emission standard for gaseous pollutants and smoke emission control.

1.3 Ways to Reduce Motor Vehicles Air Pollution

It is impossible to reduce the growth in motor vehicle use in the community as a trade off to benefit in mobility in order to reduce air pollution. It is a priority to control and manage the growth of motor vehicles usage as well as using technical measures to minimize their adverse impact on the environment and consequently human being.

1.3.1 Infrastructure and Public Transport Improvements

Appropriate design of roads, intersections, and traffic control systems can reduce bottlenecks, accommodate public transport, and smoother traffic flow at moderate cost. New roads to relieve bottlenecks and to accommodate public transport, are essential as part of an integrated plan to reduce traffic congestion and minimise urban air pollution. Similarly, strengthening public transport system will be effective to discourage private car and motorcycle use.

1.3.2 Technical Measures to Limit Vehicular Air Pollution

1.3.2.1 Noncatalytic approach

(a) Air-fuel Ratio Control

The air-fuel ratio has an important effect on engine power, efficiency and emissions. The ratio of air to fuel in the combustible mixture is a key design parameter for spark-ignition engines. An air-fuel mixture that has exactly enough air to burn the fuel is the stoichiometric ratio and has normalized air-fuel ratio (λ) of 1.0. Mixtures with more air than fuel are lean with λ higher than 1.0. Mixtures with more fuel than air are rich with λ less than 1.0. Rich mixture combustion will create more unburned hydrocarbon while lean mixture combustion will cause more NOx emission (Heck et al., 2002).

(b) Electronic Control System

Electronic control technology to maintain stoichiometric combustion has been extensively developed. The system calculates the air-fuel ratio by sensing oxygen level in the exhaust and adjusts the air-fuel mixture at stoichiometry. In addition, computer systems control spark timing, exhaust gas recirculation, idle speed, air injection systems to control emission (Heck et al., 2002).

(c) Crankcase Emission and Control

The blow-by of compressed gases past the piston rings consist mostly of unburned hydrocarbons. In uncontrolled vehicles, the blow-by gases were vented to the atmosphere. Crankcase emission controls involve closing the crankcase vent port and venting the crankcase to the air intake system via a check valve. Thus, the unburned hydrocarbons will be re-circulated into the combustion system instead of vented out to atmosphere (Heck et al., 2002).