

# Effect of Geometric Parameters on the Performance of Automotive Catalytic Converters

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## ABSTRACT

*This paper describes a parametric study to analyze the performance of three-way catalytic converters under transient conditions. The study is carried out to facilitate the design improvement of catalytic converters in order to meet the progressively tightening emission regulations. The converter performance is simulated by considering chemical reactions and heat transfer phenomena as the exhaust gases flow through the catalyst. The model also takes into account the adsorption/desorption of oxygen in the catalyst during non-stoichiometric composition of air-fuel mixtures. The discretized governing equations are solved by tri-diagonal matrix algorithm with a successive line under relaxation method. The results show that increasing the length or cross-sectional area of the catalyst increases the conversion efficiency. The effect of changing length is more important for short converters and it becomes gradually less significant for long converters. The results show that a short catalyst with a large cross-sectional area is generally not as effective in pollutant conversion as a catalyst of similar volume but of larger length and smaller cross-sectional area. The increase of cell density and decrease of wall thickness have favorable effect on catalyst conversion performance. The combination of high cell density and thin wall increases the catalyst geometric surface area, which improves the conversion performance. HC emissions are found to be more sensitive to the effect of geometric surface area.*

## 1. INTRODUCTION

Three-way catalytic converters (TWC) have been employed in automobiles to reduce the exhaust emissions for more than thirty years. They have done a tremendous job in reducing the engine emissions by more than 90%. However, with progressively stricter emission regulations, the catalytic converter design and performance need to be continuously improved. Design improvement efforts are focused on reducing the emissions during both the cold start conditions and the normal driving operations. There are various approaches for the design improvements. Some of them, such as electrically or chemically heated catalysts, are focused mainly on reducing the cold start emissions by minimizing the catalyst warm up period during the cold engine starts. Other studies are focused on improving the catalyst performance during the post warm up period. In addition to applying innovative approaches for reducing emissions, efforts are being made for design optimization of the existing three-way catalytic converters. Such efforts involve optimization of the converter geometry, selection of substrate and washcoat materials and the converter

location in the exhaust line. Due to cost-effectiveness, mathematical modeling is better suited for optimization studies.

Mathematical models of catalytic converters have been employed for nearly 30 years. Most of these models [1-7] are one-dimensional, and they consider property gradients only along the catalyst axis. Furthermore, these models generally assume a uniform flow in the catalyst and ignore the effect of pressure variations. The effect of property gradients normal to the flow is assessed by using two-dimensional [8,9] and three-dimensional models [10]. These studies show that one-dimensional models do a fairly good job in predicting the overall catalyst performance. In the existing models, the heterogeneous reactions at the catalyst substrate surface are modeled by using different chemical reaction mechanisms. The mechanisms considered range from a simple 3-step chemistry [10] to a more inclusive 13-step chemistry [5]. The oxygen storage capacity of the wash-coat material, which provides the ability to periodically store and release oxygen to improve conversion efficiency [11], is modeled by using additional reactions.

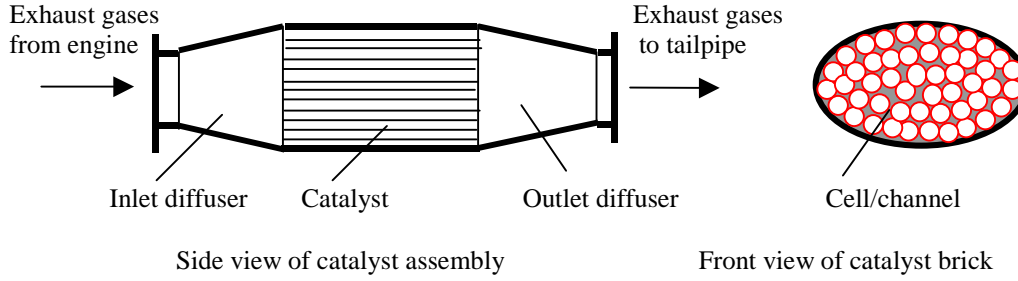


Figure 1. Schematic of a three-way catalytic converter

Most of the existing models have been developed to analyze and predict the converter performance under different driving conditions. Some studies [12] used models to simulate the converter performance during legislated driving cycles. Some other studies [7,13,14] employed models to investigate the dynamic behavior of converters. They studied the effect of oscillations in feed gas composition and air-fuel ratio, and their influence on catalyst conversion efficiency.

One of the desirable applications of mathematical modeling is in the area of design optimization. A reliable and efficient simulation may reduce the number of expensive and time-consuming experiments and provide better insight. However, a critical review of literature on catalyst modeling reveals that numerical modeling has rarely been used for converter design optimization. The present study was motivated by recognizing this gap in the literature. In this study, the effect of geometry (length, cross-sectional area, cell wall thickness and cell density) on the converter performance during the US Federal Test Procedure (FTP) is assessed.

## 2. MATHEMATICAL FORMULATION

Figure 1 shows a schematic of the catalytic converter. The governing equations were developed by considering the conservation of mass, energy and chemical species. The following phenomena are explicitly included in the model: (i) Convective heat and mass transfer from the exhaust gas to the catalytic surface; (ii) Heterogeneous chemical reactions taking place on the catalytic surface; (iii) The catalyst's capacity of storing extra oxygen under lean conditions and releasing it under rich conditions; (iv) Heat losses to the surroundings; and (v) Heat conduction along the catalytic converter. The governing conservation equations are listed below:

*The gas phase energy equation:*

$$\rho_g C_{p_g} \left( \varepsilon \frac{\partial T_g}{\partial t} + v_g \frac{\partial T_g}{\partial z} \right) = -h_g G_a (T_g - T_s)$$

*The gas phase species equations (for 7 species):*

$$\left( \varepsilon \frac{\partial C_g^j}{\partial t} + v_g \frac{\partial C_g^j}{\partial z} \right) = -k_m^j G_a (C_g^j - C_s^j)$$

*The surface energy equation:*

$$(1 - \varepsilon) \rho_s C_{p_s} \frac{\partial T_s}{\partial t} = (1 - \varepsilon) \lambda_s \frac{\partial^2 T_s}{\partial z^2} +$$

$$h_g G_a (T_g - T_s) - h_{\infty} S_{ext} (T_s - T_{\infty}) +$$

$$G_a \sum_{k=1}^{n_{reaction}} R^k (T_s, C_s^1, \dots, C_s^{n_{species}}) \cdot \Delta H^k$$

*The surface species equations (for 7 species):*

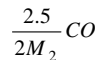
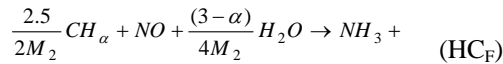
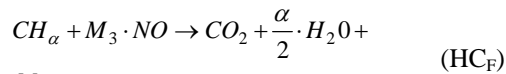
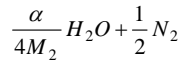
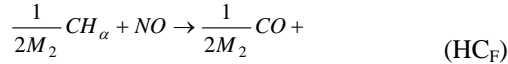
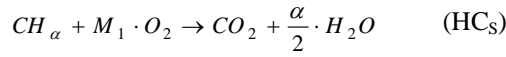
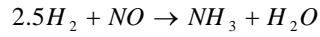
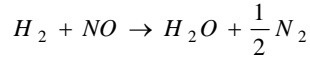
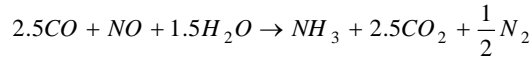
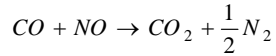
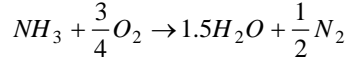
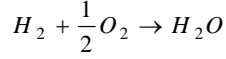
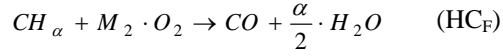
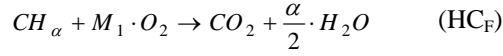
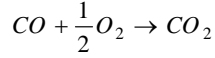
$$(1 - \varepsilon) \frac{\partial C_s^j}{\partial t} = km^j G_a (C_g^j - C_s^j) -$$

$$G_a R^j (T_s, C_s^1, \dots, C_s^{n_{species}})$$

where the superscript  $j$  varies from 1 to 7 representing, respectively, the following gas and surface species: CO, NO, NH<sub>3</sub>, O<sub>2</sub>, C<sub>3</sub>H<sub>6</sub>, H<sub>2</sub> and C<sub>3</sub>H<sub>8</sub>. The conservation equation for the surface oxygen storage mechanism can be represented by the surface species equation excluding the convective mass transport term.

The governing equations were discretized by using a non-uniform grid and employing the control volume approach and the central implicit difference scheme in the spatial direction. Since chemical reactions change composition more rapidly near the inlet, smaller grid spacing is used near the inlet and larger spacing near the exit. The equations were solved by using a standard tri-diagonal matrix algorithm (TDMA) with a successive line under-relaxation scheme. The surface chemical reactions are modeled using a 13-step reaction mechanism. This mechanism consists of 13 independent forward pathways for oxidation of CO, H<sub>2</sub>, C<sub>3</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub>, and NH<sub>3</sub> with O<sub>2</sub> and NO as oxidizing agents, and their corresponding rich and lean kinetic rate expressions. This chemical reaction scheme and kinetic data were originally presented by Otto

and LeGray [3] and later presented with the modified kinetic data by Montreuil et al. [5]. The reaction scheme is shown as follows:



where  $\alpha$  = hydrogen-to-carbon ratio, and

$$M_1 = [1 + \frac{\alpha}{4}]; M_2 = [\frac{1}{2} + \frac{\alpha}{4}]; M_3 = [2 + \frac{\alpha}{2}]$$

Here,  $HC_F$  = fast burning hydrocarbon and  $HC_S$  = slow burning hydrocarbon.

In addition to the catalyst's substrate kinetics, the model also includes an additional 9-step chemistry to describe the oxygen storage capacity of the substrate wash-coat material. A detailed description of the model, the details of the chemical mechanisms, the corresponding kinetic data, solution procedure, initial and boundary conditions are given by Shamim et al. [12].

### 3. RESULTS AND DISCUSSION

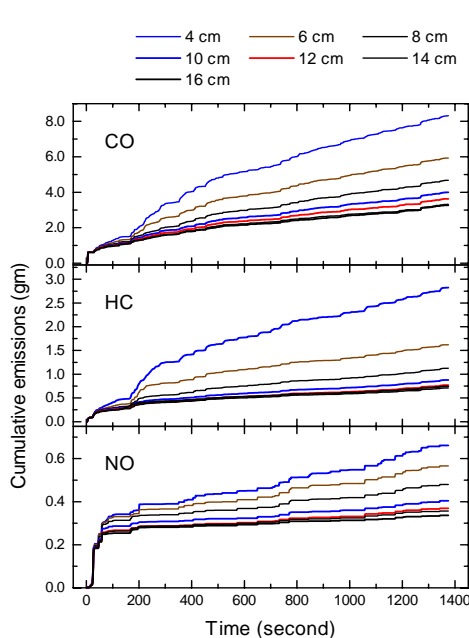
The model was employed to study the effect of converter geometry on the catalyst pollutant conversion efficiency. The actual driving conditions

were simulated by considering the converter operation during a legislated driving test cycle. The US Federal Test Procedure (FTP) was selected. During the FTP, the vehicle is driven at specified speeds for a specified time (1372 s for FTP-72) and covers a distance of 7.5-mile (12.07 km). The exhaust gases from the engine and tailpipe are measured and are collected in tedlar bags during the test cycle. This procedure yields the amounts of engine exhaust emissions in g/mile for comparison with legislated values. In the present study, the numerical model is used to calculate the cumulative emissions. In addition to the emissions during the entire legislated cycle, the catalyst cold start performance is assessed by collecting emissions during the first 140 seconds of the test. This period represents the first "hill" of the FTP cycle.

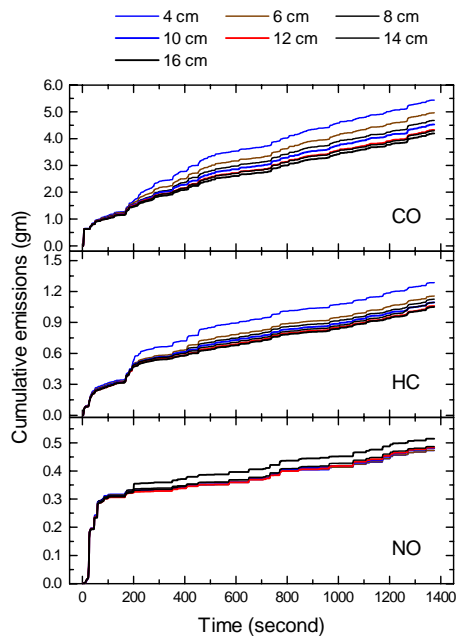
The numerical model was validated using experimental measurements at both steady and transient conditions. The model validation details are given elsewhere [12]. The converter used for the base case was a palladium-based catalyst, with a length of 8 cm, cross-sectional area of 86.054 cm<sup>2</sup>, cell density of 62 cell/cm<sup>2</sup>, and wall thickness of 0.1905 mm. The following sections present the discussion of the effect of varying different dimensions of the catalyst. These results were obtained by changing one or two parameters at a time while keeping the other parameters constant. It must be mentioned that, due to the effects of catalyst composition and aging, the trends found here may only be qualitatively generalized.

#### 3.1 Effect of the Catalyst Length

The effect of catalyst length was investigated by varying the length from 4 cm to 16 cm. Figure 2a shows the cumulative emissions as a function of time for different catalyst lengths while keeping cross-sectional area constant. The results depict, as expected, that emissions decrease with the increase of the catalyst length. This emission decrease is partly due to an increase in the reaction surface area. The effect of length becomes gradually smaller with an increase of length. For example, CO emissions drop by 43.7% when length is increased from 4 cm to 8 cm. However, the drop in CO emissions is only 30.1% when length is increased from 8 to 16 cm. The percent reduction in emission with an increase of length is different for different species. For the given operating conditions, the CO and HC emissions become insensitive to an increase in length beyond 14 cm. NO emissions, on the other hand, exhibit some sensitivity to changes in length beyond 14 cm.



(a)



(b)

Figure 2. Cumulative emissions during the FTP for different catalytic converter lengths: (a) variable catalyst volume; (b) fixed catalyst volume.

The increase of length decreases CO and HC emissions for a fixed volume case as well (see Figure 2b). However, the sensitivity of emissions to lengths is relatively small. NO emissions, on the other hand, are found to increase with an increase of length. This indicates that major part of NO conversion occurs near the catalyst inlet. Figure 3 shows the cumulative emissions during the whole FTP as a function of the converter length for both cases of fixed cross-sectional area (variable volume) and fixed volume. The figure shows that, for a short catalytic converter with a fixed cross-sectional area, an increase in the length leads to a relatively large decrease in emissions. The effect of length becomes gradually small with an increase of the length. Hence, as mentioned earlier, there does not appear to be a significant advantage of increasing the length of a long converter. This is due to an increase in the substrate thermal capacity of a large volume converter that delays the catalyst in reaching the light-off temperature and thus increases the cold start emissions. Long converters result in a slow warm-up process, which is adverse to the pollutant conversion. An increase of the catalyst volume also increases the cold start emissions as has been found by many experimental studies [15].

### 3.2 Effect of Cross Sectional Area of the Catalyst

The catalyst cross-sectional area was varied while keeping the length constant. Thus an increase in cross-sectional area represents an increase in the catalyst volume. Figure 4 shows the cumulative emissions for catalysts with different cross-sectional areas. As expected, the emissions decrease with the increase in the cross-sectional areas. This decrease in the emission is due to an increase in the reaction surface area. The emissions levels approach to constant levels at higher areas. This is due to increase in substrate thermal capacity and ambient heat losses with an increase of catalyst volume, which have unfavorable effects on emission conversion. Hence, similar to length, increasing cross-sectional area beyond certain value for any catalyst has diminishing effect on conversion performance. For fixed catalyst volume, an increase of cross-sectional area increases CO and HC emissions, and does not have much influence on NO emissions except at small values of area where NO shows a slight decrease. The results show that a short catalyst with a large cross-sectional area is generally not as effective in pollutant conversion as a catalyst of similar volume but of larger length and smaller cross-sectional area.

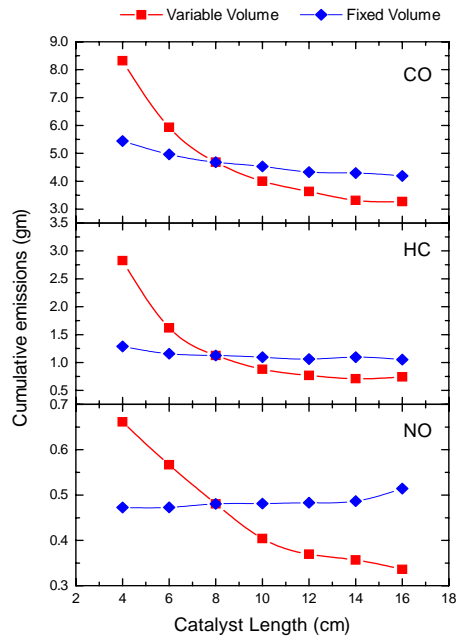


Figure 3. Effect of catalyst length on cumulative emissions for fixed and variable catalyst volume

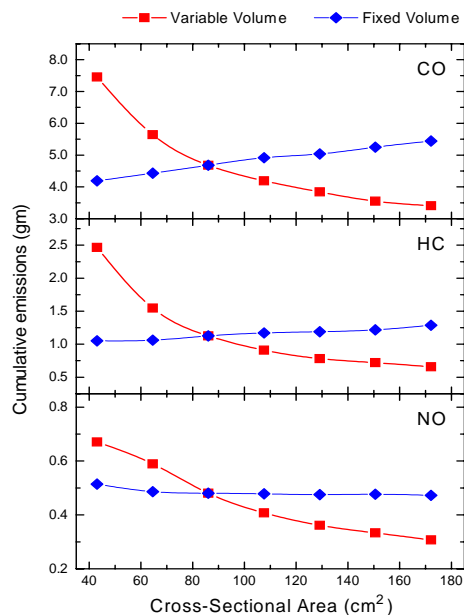


Figure 4. Effect of catalyst cross-sectional area on cumulative emissions for fixed and variable catalyst volume

Figure 5 compares the effect of length and cross-sectional area on catalyst performance for different volumes. For variable length case, the cross-sectional area was kept at 86.054 cm<sup>2</sup>, and for variable area case the length was kept at 8 cm.

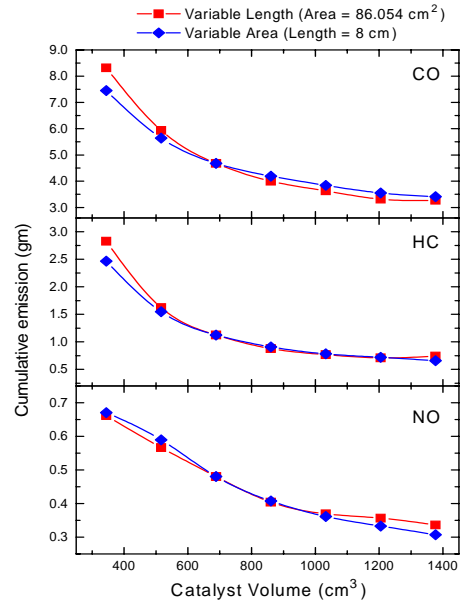


Figure 5. Comparison of the effect of catalyst length and cross-sectional area on the catalyst performance

The figure shows that increase of catalyst volume by increasing either the length or cross-sectional area has favorable effect on emission reduction. However, the pollutant conversion is relatively more sensitive to changes in length than the cross-sectional area. For the given catalyst geometry, the reduction in catalyst volume by reducing the converter length results in much higher CO emissions than that obtained by corresponding reduction in converter cross-sectional area. Similarly, the increase in catalyst volume by increasing the length results in lower CO emissions than that obtained by corresponding increase in the catalyst cross-sectional area. HC emissions show similar trends as CO at low volumes. For higher catalyst volumes, the effects of length and cross-sectional area on HC emissions are found to be similar. Contrary to CO and HC emissions, NO emissions show relatively greater sensitivity on cross-sectional area than the catalyst length. NO emissions are slightly higher for longer catalysts with smaller cross-sectional area than catalysts of similar volume but larger cross-sectional area and shorter length.

### 3.3 Effect of Monolith Cell Density and Wall Thickness

Several experimental studies have shown that the converter performance can be improved by reducing the bulk density and increasing the geometric surface area of the substrate [15-17].

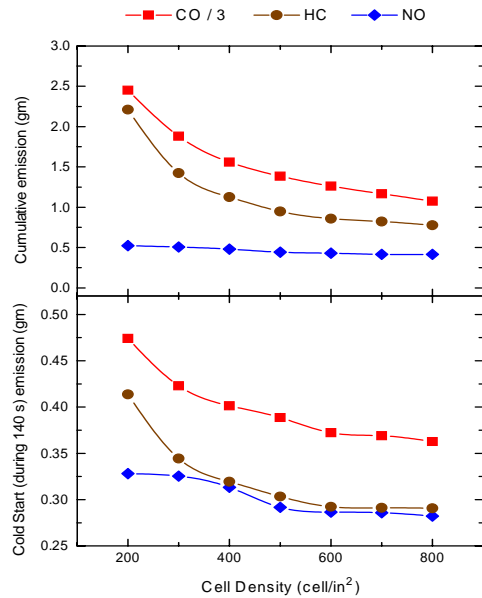


Figure 6. Effect of catalyst cell density on the catalyst performance

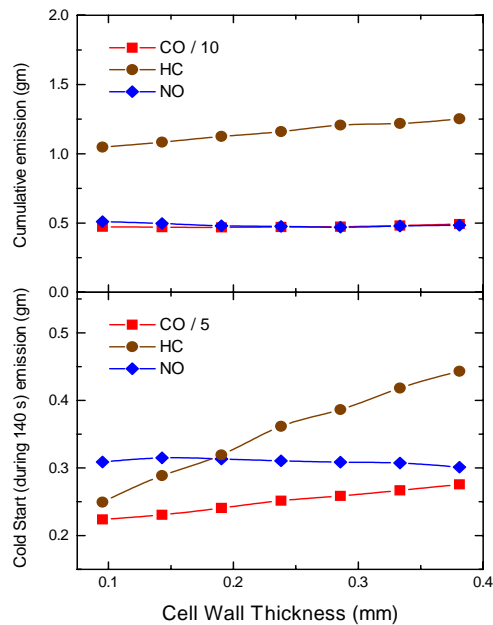


Figure 7. Effect of catalyst cell wall thickness on the catalyst performance

This may be achieved by using converters with high cell density and low wall thickness. Early monolithic

converter had cell density of 46.5 cell per square centimeter (cpscm) (300 cell per square inch - cpsi) with a wall thickness of 0.31 mm (12 mil) [18]. Advancement in technology has allowed the production of converters with thinner wall thickness and higher cell density. In modern automobiles, converters with 0.11 mm (4 mil) wall thickness and 62 cpscm (400 cpsi) are commonly used. Studies have been carried out for the feasibility of employing 0.06 mm (2 mil) wall thickness with 139.5 cpscm (900 cpsi) [15].

Figure 6 depicts the effect of increasing cell density on pollutant emissions. As expected, the cold start and overall emissions decrease with an increase of cell density. This decrease, as mentioned earlier, is due to a higher surface reaction area per unit volume. Compared to NO emissions, CO and HC emissions are found to be influenced more significantly by cell density. The effect of cell density is much greater for cell densities lower than 400 cpsi. NO emissions, on the other hand, remain pretty much constant for cell densities ranging from 200 to 800 cpsi. However, the cold start NO emissions decrease with an increase of cell density. The cold start CO and HC emissions also decrease with an increase of cell density, in similar pattern as the total emissions during the entire FTP cycle. It should be noted that the higher value of cell density is limited by the consideration of the corresponding increase in back pressure (the engine exhaust pipe pressure). If the back pressure is too high, it will deteriorate aspiration of the engine and, thereby, the engine performance. To avoid this, a thinner wall substrate is necessary for a higher cell density.

Figure 7 shows the effect of substrate wall thickness keeping other parameters (especially the cell density) constant. The result shows that HC emissions increase with an increase of wall thickness. The increase in emissions for thicker wall is due to a corresponding decrease in geometric surface area (GSA). However, the CO and NO emissions are not much influenced by a change of wall thickness in the range of thickness studied. NO emissions show a slight decrease with an increase of wall thickness, which is due to an increase in NO reaction with the corresponding higher amount of HC present. The wall thickness has much greater influence on cold start emissions. Both CO and HC cold start emissions increase with wall thickness. This increase in emissions for thicker walls is due to a corresponding increase in the substrate heat capacity, which increases the time for the converter to reach its light-off temperature. The cold start NO emissions, however, decrease for thicker walls, which is due to a corresponding increase of NO reaction with higher amount of HC present.

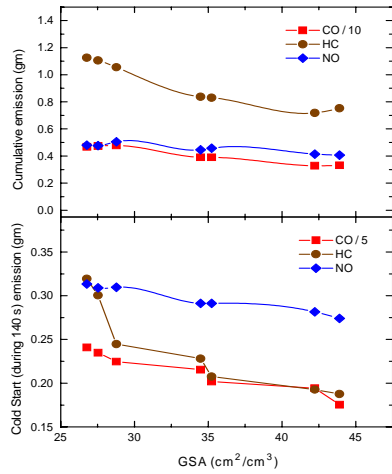


Figure 8. Effect of geometric surface area (GSA) on the catalyst performance

A thinner wall substrate also makes it feasible to use higher cell density. Umehara et al. [15] used various substrates of different combinations of wall thickness and cell density to experimentally study the effects on pollutant emissions. In the present study, simulations were carried out by selecting six different sets of wall thickness and cell density. The wall thickness / cell density are 0.1905 mm (7.5 mil) / 62 cpscm (400 cps), 0.16 mm (6 mil) / 62 cpscm (400 cps), 0.11 mm (4 mil) / 62 cpscm (400 cps), 0.11 mm (4 mil) / 93 cpscm (600 cps), 0.09 mm (3 mil) / 93 cpscm (600 cps), 0.09 mm (3mil) / 139.5 cpscm (900 cps), and 0.06 mm (2 mil) / 139.5 cpscm (900 cps) respectively. As the wall thickness decreases and the cell density increases, the geometric surface area (GSA) increases. Figure 8 shows the emissions as a function of GSA. The results show that pollutant emissions generally decrease with the increase in GSA. The reduction is greater for HC emissions. When the GSA is increased from 26.77 cm<sup>2</sup>/cm<sup>3</sup> to 43.90 cm<sup>2</sup>/cm<sup>3</sup>, the total FTP and the cold start HC emissions decrease by 33.1% and 41.2% respectively. For a similar increase of the GSA, the decrease in the total FTP and the cold start CO emissions are 29.2% and 27.2% respectively. The effect of increase in GSA on NO emissions is much less. This trend is similar to the experimental results of Umehara et al. [15] even though the engine used for the present study is different from that in this reference.

#### 4. CONCLUSIONS

- A numerical model for simulating the performance of three-way catalytic converters was employed to investigate the effect of various

catalytic converter geometric design parameters on converter performance under transient driving conditions.

- The conversion efficiency of catalyst increases with the increase of catalyst length. The change in length is more important for short converters and it becomes gradually less important for long converters. Keeping other parameters constant, increasing the length of long converters may even have an adverse effect on the conversion efficiency.
- For a fixed catalyst volume, the increase of length decreases CO and HC emissions but increases NO emissions. The increase of cross-sectional area for fixed catalyst volume (achieved by reducing length), however, increases CO and HC emissions, and does not have much influence on NO emissions. The results show that a short catalyst with a large cross-sectional area is generally not as effective in pollutant conversion as a catalyst of similar volume but of larger length and smaller cross-sectional area.
- The increase of cell density and decrease of wall thickness have favorable effect on catalyst conversion performance. The combination of high cell density and thin wall results in an increase in substrate geometric surface area and has favorable effect on the catalyst emission conversion performance.

#### ACKNOWLEDGMENTS

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#### NOMENCLATURE

$C_g^j$	gas phase concentration of species $j$ , mol/m <sup>3</sup>
$C_s^j$	surface concentration of species $j$ , mol/m <sup>3</sup>
$c_{pg}$	specific heat of gas, J/kg·K
$c_{ps}$	specific heat of substrate, J/kg·K
$G_a$	geometric surface area, m <sup>2</sup> /m <sup>3</sup>
$\Delta H^k$	heat of reaction of species $k$ , J/mol
$h_g$	heat transfer coefficient between flow and substrate, J/m <sup>2</sup> ·s·K
$h_\infty$	heat transfer coefficient between substrate and atmosphere, J/m <sup>2</sup> ·s·K
$k_m^j$	mass transfer coefficient for species $j$ , m/s
$R^k$	reaction rate of $k^{\text{th}}$ reaction, mol/m <sup>2</sup> ·s
$S_{ext}$	external surface to volume area ratio, m <sup>2</sup> /m <sup>3</sup>
$t$	time, s
$T_\infty$	ambient temperature, K
$T_g$	gas temperature, K
$T_s$	substrate temperature, K
$v_g$	gas flow velocity, m/s

$z$	coordinate along catalyst axis, m
$\varepsilon$	void volume fraction
$\lambda_g$	thermal conductivity of gas, J/m·s·K
$\lambda_s$	thermal conductivity of substrate, J/m·s·K
$\rho_g$	gas density, kg/m <sup>3</sup>
$\rho_s$	substrate density, kg/m <sup>3</sup>

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