EXPERIMENTAL ANALYSIS AND COMPARISON OF PERFORMANCE CHARACTERISTICS OF CATALYTIC CONVERTERS INCLUDING SIMULATION

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ABSTRACT
The purpose of this paper is to present the results of an experimental study of the performance and conversion efficiencies of ceramic monolith three-way catalytic converters (TWCC) employed in automotive exhaust lines for the reduction of gasoline emissions. Two ceramic converters of different cell density, substrate length, hydraulic channel diameter and wall thickness were studied to investigate the effect of varying key parameters on conversion efficiencies and pressure drop. Based on the emission test results, the conversion efficiencies from both converters were calculated and evaluated. After completing the test, the converters were cut to extract the substrate or ‘honeycomb’ inside the housing and being analyzed for microstructure and materials composition using Scanning Electron Microscopy (SEM) and Energy Dispersive Analysis (EDX).

Simulation program using commercial computational fluid dynamics (CFD) software packages, GAMBIT and FLUENT 6.1 was used to verify experimental results.

Keywords: Catalytic converter, Exhaust emission, Conversion efficiency, Substrate, Energy dispersive analysis.

INTRODUCTION
A serious issue that is always been debated among the environmentalists over the decades and recent years is air pollution. As the technology keep on evolving and emerging, it carries along undesirable effects apart from its broad application and use. One of the main contributors is said to be the emission of harmful gases produced by vehicle exhaust lines. The number of vehicles miles travels per year continues to increase as a result of higher demand and needs. Consequently, an increase in the number led to the increase of the content of pollutants in air.

The need to control engine emissions was recognized as early as 1909. Due to the more stringent rules and emission standards, automotive manufacturers began to develop a treatment device for exhaust gases known as catalytic converter for their vehicle models. General Motors (GM) was the pioneer in the early 1970s followed by Ford and Chrysler. Catalytic converters came in many concepts, structures and even the materials; nevertheless, it continued to evolve depending on different vehicle requirements.

The conversion process is performed by means of catalyst which accelerates the chemical reactions. It remains unchanged through the process and able to sustain high temperatures caused by incoming exhaust stream. Most frequently, precious metals such as Platinum (Pt), Palladium (Pd), Rhodium (Rh) and Vanadium (V) are being used as catalysts and because of their rareness and outstanding ability, catalytic converters become among the most expensive devices in a vehicle. Though the researchers begin to replace them with oxides of base metals, which are much cheaper, such as Zinc (Zn), Aluminum (Al) and Magnesium (Mg), however, due to their lower performance compared to the precious ones, they do not have any other choice rather than to keep on implementing those expensive metals for automotive catalysts and other industrial applications.

The experiment is carried out to analyze the performance characteristics and behavior of the three-way ceramic monolith catalytic converter (TWCC) especially its efficiency in reducing the amount of pollutants. Experiment is conducted to compare the performance of two ceramic converters of different hydraulic diameter, channel length and cell density on conversion efficiencies and pressure drop. By observing the results, suggestions on the considerable design geometries for catalytic converter properties are made.

Emission test on engine test bed with converter installed and how engine operating conditions would influence the performance of catalytic converter is investigated. The test is done at various engine speeds and at different air-to-fuel ratio.

The content of carbon dioxide (CO₂), carbon monoxide (CO), hydrocarbon (HC), oxides of nitrogen (NOₓ) and oxygen (O₂) are measured before and after the converters using the MRU Delta 1600-L multi-gas analyzer which is used during the emission test.

After completing the test, the converters were cut to extract the substrate or ‘honeycomb’ inside the housing and being analyzed for microstructure and materials composition using Scanning Electron Microscopy (SEM) and Energy Dispersive Analysis (EDX).

The main objective of the paper is to compare and analyse the performance characteristics of converters from PROTON Wira 1.3L and another one from FIAT Punto Selecta 1.2L. Both converters had same substrate material.

To verify the results obtained from the experimental works, computer simulation of a catalytic converter system is designed.
where catalyst reactions take place. Based on the converter parameters and material properties, the mass fraction can be observed graphically with the aid of computational fluid dynamics software, FLUENT 6.1. This enables a “clearer” view on how these parameters vary with channel geometries.

CATALYTIC CONVERTERS
Catalytic converter has gone through many processes and remarkable evolution for the past 30 years. It is said to be one of the most effective tool to fight against the overwhelming pollutant contents in our environment, as it reduces almost 80% of the harmful gases resulting from the incomplete combustion of the engine. Catalytic converter is a stainless steel container mounted somewhere along the exhaust pipe of the engine and inside the container is a porous ceramic structure through which the exhaust gas flows (Ganesan, 2004). In most converters, the ceramic is a single honeycomb structure with many flow passages. The passages comprise of many shapes, including square, triangular, hexagonal and sinusoidal. Early converters used loose granular ceramic with the gas passing between the packed spheres. Since it is difficult to keep the spheres in place, many converter developers opted for ceramic monolith which offers various advantages. Among these advantages are smaller volumes, lower mass and greater ease of packaging (Heck & Farrauto, 1995).

The active catalyst layer is applied on the monolith walls. The coating, called ‘washcoat’, is composed of porous, high surface area inorganic oxides such as γ-Al₂O₃ (gamma alumina), CeO₂ (Ceria) and ZrO₂ (Zirconia). Noble metal catalysts, such as Platinum (Pt), Palladium (Pd) and Rhodium (Rh), are deposited on the surface and within the pores of the washcoat (Pontikakis, 2003). Exhaust gas flowing in a catalytic converter diffuses through the washcoat pore structure to the catalytic sites where heterogeneous catalytic reactions occur. The washcoat layer on metallic foil and on ceramic substrate is illustrated in Figure 1. The thickness of the washcoat layer is typically 20-100 μm. Much thicker washcoat deposits (“fillets”) are formed in the cell corners, especially in the sinusoidal channels of metallic substrates.

Figure 1: Washcoats on metallic and ceramic substrates

Before the three-way catalytic converter (TWCC), two-way converters or so called oxidation catalysts were widely used in the automotive industry for carbon monoxide and hydrocarbon oxidation. Reduction of noxious gas (NOₓ) was relaxed, until 1979 where TWCC began to be implemented in automotive exhaust systems of majority vehicle models (Heck & Farrauto, 1995). TWCC provides both oxidation and reduction processes to replace two-way converter and generally, the CO reaction begins first, followed by the HC and NOₓ reactions. With lean-mixture conditions the catalyst promotes the complete oxidation of HC and CO:

\[
2CO + O_2 \rightarrow 2CO_2
\]

With rich-mixture operation, the catalyst promotes the reduction of NOₓ by reactions involving HC and CO:

\[
4HC + 10NO \rightarrow 4CO_2 + 2H_2O + 5N_2
\]

When TWCC is used, it requires an engine management system capable of very accurate air/fuel ratio control, and a catalyst. Actually, the emissions of CO, NOₓ and HC vary between different engines and are dependent on such variables as ignition timing, load, speed and, in particular, fuel/air ratio.

Latest works and research papers emphasized more on predicting the performance of catalytic converter in an early design stage, rather than setting up an experiment or laboratory test bench. Joachim Braun et al. (2002) mentioned that for the three-way catalyst, the major concern is the correct prediction of light-off behavior of the exhaust system. By modelling a converter with proper dimensions and its related properties, it is possible to see the situation that happens during the complicated chemical processes. In order to achieve reliable results, the numerical simulations have to be based on accurate models of all the significant chemical and physical processes in the catalytic converter.

The latest computational fluid dynamics (CFD) software utilized in the catalytic converter analysis is the newly developed DETCHEM³MONOLITH, acronym of ‘Detailed Chemistry’ used for the numerical simulation of the transient behavior of the TWCC. The code combines the two-dimensional simulations of the reactive flows in a representative number of monolith channels with a transient simulation with the three-dimensional temperature field of the solid structure of the converter including insulation and canning (Braun et al. 2002). The chemical reactions are modelled by a multi-step heterogeneous reaction mechanism, which is based on the elementary processes on the platinum and rhodium catalysts used.

FLUENT is also CFD software used for the same purpose. The model is constructed and meshed first in GAMBIT, a drafting program bundled with FLUENT and the finished model is then exported to FLUENT for further analysis. Catalytic converter substrate is defined as a porous media in FLUENT, where viscous and inertial losses are specified in both the streamwise and transverse directions. By using the porous media model, the number of cells in the computational mesh can be reduced significantly, since the small geometric details of the substrate do not need to be resolved.
Apart from simulating flow in catalytic converter, another method to optimize the converter performance is through the geometry modification and analysis. Substrate geometries have a great influence in catalytic activity as well as the flow pattern in the channel. The cell shape influences some parameters such as thermal mass, geometric surface area, and washcoat distribution (Presti et al. 2002). Per Marsh et al. (2001) have shown that less favourable mass transfer and higher flow velocity can be found as a results of the reduction of diameter, however, compensated with the cases of lower cell densities, implemented on both converters for separate reduction and oxidation purposes. Single heat shield is applied to Fiat converter while double heat shields that cover both the upper and bottom part are used in Proton converter. Also, for each converter, the inlet and outlet cones were fabricated identically.

The engine used in the test is a spark-ignition (inline 4 cylinder) Proton engine coupled with eddy-current type dynamometer. It is controlled by AUTOTEST VI main controller located outside of the engine room with the aid of AUTOTEST VI computer software via numerous sensors. The gas analyzer employed in the setup was the upgraded version (MRU Delta 1600-L) which is capable to measure simultaneous reactions of carbon monoxide, carbon dioxide, hydrocarbon, oxygen and also nitrogen oxides. It also incorporates a probe, suitable to measure the gases at the inlet and outlet region, along with the thermocouple, to attain temperature values for ambient and the exhaust gas itself. Figure 2 shows the complete experimental setup.

The substrate materials, type of catalysts and their corresponding percentage loading for both converters were determined using the EDX machine that is coupled with the scanning electron microscopy (SEM) system.

![Figure 2: Experimental Setup](image-url)
RESULTS AND DISCUSSION

The objective of the experiment was to observe the behavior of the catalytic converters from cold condition. Usually, this happens about 2 minutes after engine starts and based on this, a transient analysis can be built up to show the variation of main pollutants content over a period of time. However, for this analysis (mainly at idling condition), this range of temperature cannot be reached. As a result, the pollutants reduction or gas conversions were absent. For that reason, a set of recommended transient analysis for the experiment was then transferred to steady-state analysis, where the temperature was risen up to 250°C by increasing the speed and then set back to idling mode to ensure efficient converter operation. However, for uniformity, the cold start consideration was eliminated for both experiment and analysis. The tabulated data were plotted into graphs to clearly indicate the composition of pollutants before and after passing through the catalytic converter over half an hour duration.

Figure 3: CO contents in Proton Converter

Figure 4: CO contents in Fiat Converter

Figure 5: HC contents in Proton Converter

Figure 6: HC contents in Fiat Converter

Figure 7: NO contents in Proton Converter

Figure 8: NO contents in Fiat Converter
Figures 3 – 8 show the variation of composition of pollutants before and after the converters for both Proton and Fiat converters with speed. All the figures show an increase of pollutants as the speed increases except in the case of carbon monoxide for Proton converter, which has the highest amount at idling speed of 1000 rpm with a range of 4.0 to 4.5% by volume. Theoretically, due to the incomplete combustion of rich mixture, higher concentration of CO is produced. Meanwhile, the amount of CO after the converter at the same speed maintains the highest level among all speeds with a range between 2.0 to 2.5% by volume. Fiat converter performed much better in oxidizing the CO with a lower range of overall percentage, i.e., around 0.5 to 2.0% before converter and 0.0 to 1.25% after the converter. Experimental data show that the Fiat converter has higher air-fuel ratio (leaner mixture) compared to Proton converter which results the lower concentration of CO because of greater presence of oxygen.

Unburned hydrocarbon contents at idling speed of 1000 rpm is much higher in Proton converter than in Fiat converter, though the exhaust temperatures for both are similar. The hydrocarbon oxidation is still weak even kept for 30 minutes while Fiat converter has performed a HC decrease from the very beginning. It has maintained the HC level below 500 ppm for the three different speeds after passing through the substrates and again, the air-fuel ratio is one of the factors contributing to those results. For NOx reduction, both converters perform in similar way by putting the residual NO as low as 5 to 15 ppm.

**Conversion Efficiencies**

Conversion efficiency is a measure of how efficiently a converter substrate is able to reduce the amount of pollutants by means of oxidation and reduction processes. To some extent, the conversion efficiency determines the overall performance of a catalytic converter during normal or extreme conditions of the engine. The efficiency varies with many factors, including flow distribution of the exhaust gases, substrates properties, channel diameter, channel length, precious metal loading, cross-sectional area and so on. When a converter in good working order is operating at a fully warmed temperature of 400°C or above, it will remove 98-99% of CO, 95% of NOx, and more than 95% of HC.

The conversion efficiency can be further represented graphically to observe the relative change at different speeds. The catalyst is chemically inactive as engine starts and gradually active as temperature rises or when light-off temperature is achieved. Figures 9 through 11 show the comparison of HC conversion efficiencies at different rpm for both the converters.

**Scanning Electron Microscopy (SEM) Analysis**

Scanning Electron Microscopy was used for high magnification imaging and elemental analysis. A Jeol JSM-6400 scanning electron microscope equipped with an Energy Dispersive Spectrometer (EDS) was used. Flat pieces of each converter substrates were cut and placed on the specimen plate with top view directed to the microscope. The accelerating voltage and current in the measurements were 20kV and 2nA respectively, and the magnification was of 65 times. Both Proton and Fiat converters had undergone the analysis and the results are shown in Table 1.

<table>
<thead>
<tr>
<th></th>
<th>Proton converter</th>
<th>Fiat converter</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cell Density (cells/in^2)</td>
<td>400</td>
<td>600</td>
</tr>
<tr>
<td>Hydraulic Diameter (mm)</td>
<td>1.14</td>
<td>0.98</td>
</tr>
<tr>
<td>Uncoated Wall Thickness (mm)</td>
<td>0.15</td>
<td>0.12</td>
</tr>
<tr>
<td>Washcoat Thickness (μm)</td>
<td>25</td>
<td>25</td>
</tr>
</tbody>
</table>

The physical properties obtained in the analysis were used to determine other parameters that cannot be obtained experimentally such as geometric surface area, open frontal area and cell pitch.

**Electron Dispersive (EDX) Analysis**

Since both converters incorporate double substrate for separate oxidation and reduction processes, the analysis was done separately for all four substrates to examine the catalysts composition and material properties. From EDX analysis, it was found that there are different loading of precious metal catalysts at the front and rear substrates. Both converters apply the same arrangement of substrates where the front one is used for reduction of NOx while the rear one is fixed for oxidation of HC and CO. Three precious metals were found to be the catalysts are Platinum (Pt), Palladium (Pd) and Rhodium (Rh).

It was noticed that both converters have more concentrated Pd / Rh combination with small amount of Pt at the front substrate and well distributed amount of those three at the rear substrate. The major element for both front and rear substrates is Pd followed by Rh and lastly Pt. Recently, Pd usage is enhanced to replace the Pt catalyst as Pd is less expensive than Pt and more resistive to sulphur in the exhaust gas which may cause catalyst poisoning.

![Figure 9: HC conversion efficiencies at idling speed to 13000 rpm](image)
SIMULATION

For a three-way catalytic converter (TWCC), it is preferable to run simulation based on its light-off performance. Hence, the variation of temperature distribution can be clearly observed to estimate the maximum temperature achieved as a function of time. Due to experimental constraint, the objective was to compare the mass fraction values obtained from the experiment with simulation at steady state condition. At the end, the mass fraction values obtained from simulation are useful to optimize the converter geometries for maximum efficiency. For this purpose, GAMBIT and FLUENT, which came in one package, are used for both modelling and analysis.

The model was outlined in x-y coordinate system featuring all necessary points including its inlet, cone and the body. These points are related to the actual dimensions of Fiat converter such as substrate length and diameter, as well as the inlet and outlet diameters. Since the converter is symmetry, only the upper half was modelled, however, in order to visualize the full-rendered converter, the lines are connected to
each other and revolved about positive x-axis, producing a 3-D converter with a total of 98235 meshed cells (Figure 12).

After successfully modelling and meshing the converter, it was then exported to FLUENT for exhaust gas species analysis. In FLUENT, the substrate was defined as porous media, along with standard wall function treatment, meaning that the reactions with wall surface are included.

Comparison of experimental and simulation results were performed at different rpm. A typical comparison is shown in Figures 13 and 14.

CONCLUSION

Based upon the experimentation and simulation, it is concluded that
1. Fiat converter is better than Proton converter in terms of conversion efficiencies of three main pollutants CO, NO and HC at three different engine speeds. Factors affecting the performance include the geometric design of monolithic channel such as hydraulic diameter, channel length, cell density and wall thickness. Fiat converter with higher cell densities, longer channel, thinner wall and smaller hydraulic diameter produced lower emission than Proton converter.
2. Fiat converter with higher cell density and slightly thinner wall thickness allows more precious metal catalysts to be loaded hence increasing reaction surface areas for better conversion efficiency.
3. Longer channel in Fiat converter offers an advantage of better mass transfer because more exhaust gases can be attracted to the channel wall for a longer time, but at the same time produces higher pressure drop. HC and CO emission decreases as channel length increases, however, not much effect on NOx emission.
4. Smaller hydraulic diameter in Fiat converter channel exhibits more favourable mass transfer as a result of higher flow velocity. Fresh exhaust gases are easily attracted to the channel wall with small diameter to undergo chemical reactions with catalysts compared to the channel with large diameter.
5. Scanning electron microscopy (SEM) and electron dispersive analysis (EDX) showed that the properties of both converters can be compared in terms of microstructure and geometric dimension.
6. The simulation and experimental results exhibited small differences in reducing main three pollutants HC, CO and NO. It may be caused by many factors and among them is the model itself. In actual converter, double substrates were used while in simulation, only single substrate was used. Also, the precious metals cannot be arranged together in ratios in the simulation which also lead to the difference in end results.

REFERENCES