Design, analysis and fabrication of lanthanum strontium manganite catalytic converter

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Abstract. With the ever-increasing pollution rate, we have considered the need to control automobile emissions which are responsible for global warming. In this paper, we aim to propose a cost effective and sustainable design prototype of a catalytic converter. Design was done by considering some of the problems in existing catalytic converter like back pressure, cold start emissions and cost. The new design of the Catalytic converter includes a new catalyst i.e. Lanthanum Strontium Manganite (LSM) with a piecewise placement approach and a hexagonal random cell shape which is in contrast with the existing platinum-palladium catalyst and honeycomb structure. Analysis of the design of Catalytic converter was done which included the ANSYS CFD and mode shape analysis. The cold start emissions were reduced by using a method to pre - heat the air at the input of the converter shell using Arduino controlled heat plug. The testing of the converter assembly was done on a single cylinder Briggs & Stratton Engine and results were taken using Exhaust Gas Analyser which gave us the emission outputs with and without catalytic converter at idling and full rpm of the engine.

1. Introduction
A catalytic converter is a device used to control vehicle emissions by carrying out catalysed chemical reaction with by-product toxic elements from Internal Combustion engine and converting them into less toxic substances. The toxic by-products from the IC engine contains Carbon Monoxide (CO), Hydrocarbons (HC) and oxides of Nitrogen (NO\textsubscript{x}). CO and HC emissions occur as the engine start and NO\textsubscript{x} emissions are formed at higher temperature.

The catalytic converter consists of the substrate which is usually a ceramic or alumina silicate which is used to give a large surface area, for the reaction to happen. The catalyst is coated over the substrate on which the mass diffusion takes place. As the emission molecules are diffused over the catalyst, they react with the oxygen molecules trapped by the catalyst to carry out oxidation of CO and HC. The catalyst used are mostly precious metals like platinum, palladium and rhodium etc. where platinum is used in petrol engines for oxidation of CO and HC. Palladium is a choice for oxidation catalyst in diesel engines and rhodium is the reduction catalyst to reduce the NO\textsubscript{x} emissions[1]. The substrate is packaged into an insulation mat which is wrapped around the catalyst so that the heat cannot escape the catalyst area. It acts as an insulation shield as the catalyst require a constant temperature to operate. Following reactions takes place in the catalytic converter.

\textit{Oxidation of carbon monoxide to carbon dioxide}

\[2 \text{CO} + \text{O}_2 \rightarrow 2 \text{CO}_2\]
Oxidation of unburnt hydrocarbons (HC) to carbon dioxide and water,

\[ C_xH_y + 2xO_2 \rightarrow xCO_2 + 2xH_2O \]

Reduction of nitrogen oxides to nitrogen (N\(_2\))

\[ 2CO + 2NO \rightarrow 2CO_2 + N_2 \]

\[ 2H_2 + 2NO \rightarrow 2H_2O + N_2 \]

Hydrocarbon emissions is one of the hazardous emissions. Hydrocarbon emissions produce ground level ozone (produced when two pollutants react in sunlight i.e. NO\(_x\) and hydrocarbons), which causes skin cancer and eye irritation. Hydrocarbon emissions are highest during cold start, which means that in this phase, the catalyst does not reach the light off temperature or minimum temperature to start the reaction which leads to ejection of the toxic hydrocarbons directly into the atmosphere.

Anupam Mukherjee et al. [2] has shown that the incomplete combustion of the fuel in the IC engine results in the production of gaseous emissions like CO, HC and NO\(_x\) in great concentration which can damage the environment. Krunal P. Shah et al. [3] have shown that an experimental study on the use of ZrO\(_2\) coated wire mesh is being done on a 4-cylinder 4 stroke diesel engine. Parameter like backpressure and HC emissions were monitored. E. Salehi, M et.al. [4] presented results of reduction in oxides of nitrogen from exhaust gas in the presence of NH3 as reductant on selective catalytic reductant (SCR) impregnated catalytic converter. Y. Putrasari et.al. [5] presents Increasing the roughness and surface area of FeCrAl substrate using ultrasonic treatment at 35kHz frequency with SiC-methanol in order to increase the reaction rate inside the converter. Ivan Cornejoe et.al. [6] presents a RANS based computational model of turbulence decay analysis inside the catalytic converter monolith. Manish kumar Parmar et al. [7] presents Wire mesh substrate coated with SS-304,Gold and silver respectively, were tested. The wire mesh arrangement accounted for better decreased emission rates, backpressure, availability and cost. There have been many studies on backpressure of the catalytic converter both numerical and experimental for different inlet cone angles [8], hexagonal cell shapes [9] and even an aftermarket design. An increase in exhaust backpressure decreases the concentration of NO\(_x\), due to the increased exhaust gases in the cylinder. Excessive backpressure in the exhaust system creates the problem of overheating, lowers engine power output and fuel consumption in the engine cylinder, that may cause poor performance (e.g. reduced intake manifold boost pressure, cylinder scavenging and combustion effect) and damage of the engine parts (e.g. turbocharger problems – seals, increased pumping work etc.). Therefore, backpressure in a certain level (specified by the engine manufacturer) contributes to improvement of the engine performance and reduces emissions[10]. Moreover, cold start as a problem in automotive catalytic converter is recognised and methods like thermal energy storage and preheating is employed in [11,12]. The use of LSM in SOFCs as cathode material to trap oxygen is evidenced by many available literature [13-15].

2. Design and Analysis

The catalytic converter consisted of an inlet pipe with air pre heating heat plug mechanism which is Arduino controlled that operates for 15 seconds as the engine starts. Material of Inlet pipe is Stainless Steel 306 that can withstand temperatures upto 800°C. This pre-heat mechanism is used to reduce cold start hydrocarbon emissions as the catalyst in the catalytic converter is required to reach a minimum light off temperature to start the reactions.
Design of the catalytic converter is done in order to reduce backpressure which is primarily governed by inlet fulcrum design and substrate cell structure. First step is to design inlet fulcrum which is designed in Solidworks and analysed in ANSYS Fluent 18.1 to reduce the backpressure by choosing the fulcrum angle at 45 degrees that was the most optimal design angle of the fulcrum.

The catalyst shell was also SS 306 and the shell volume was calculated using the swept volume and number of intakes by the engine.  
Converter volume = V.F.R/S.V  
V.F.R = Swept volume × Number of intakes per hour  
= 0.000305 × 1900 × hr  
= 34.77 m$^3$/hr  
Density = 1.43 Kg/m$^3$  
Mass Flow Rate = 47.63 Kg/hr  
Space velocity = 34.77/0.00065 = 53492.30 hr$^{-1}$  
Substrate dimension = Dia54 X L100 m  
Substrate volume = 915.42cm$^3$

| COMPONENT                | MATERIAL                        |
|--------------------------|---------------------------------|
| INLET FLANGE             | FE410                           |
| INLET PIPE               | SS 306                          |
| INLET CLAMP FLANGE       | Mild Steel                      |
| CONVERTER SHELL          | SS 306                          |
| INSULATION MAT           | Ecoflex 400                     |
| SUBSTRATE                | Alumina coated Polyurethane     |
| OUTLET CLAM SHELL        | Mild steel                      |
| OUTLET PIPE              | SS 306                          |
| ALL CONES                | SS 306                          |

The catalyst inside the shell was supported and insulated by Ecoflex 400 mat which does not let the heat to escape from the shell and the catalyst.
2.1. CFD

Geometry of the catalytic converter is prepared in Solidworks and was simplified for mesh adjustment and high mesh quality. ANSYS Fluent solver is used to carry out the simulation. FLUENT uses finite volume method which means that it divides the whole geometry into small volumes in order to simulate over the complete computational domain.

Air is used as fluid media, which is assumed to be steady and compressible. Flow simulations were performed at steady state through the catalytic converter using ANSYS 18.1. The flow inside the substrate was treated as laminar due to very small hydraulic diameter of the flow channels. The volume of flow domain was modelled by standard k - ε turbulence model. The governing equations for the flow within the domain are the Reynolds Averaged Navier-Stokes (RANS) equations. The total mass conservation equation (continuity equation), is

\[ \frac{\partial \rho}{\partial t} + \nabla (\rho \mathbf{v}) = 0 \]

\( \rho \) in kg /m\(^3\) - density; \( t \) (seconds)- time; \( \mathbf{v} \) in m/s - fluid velocity vector.

The momentum conservation equation is,

\[ \frac{\partial}{\partial t}(\rho \mathbf{v}) + \nabla (\rho \mathbf{vv}) = -\nabla p + \nabla \tau + \rho g + S_i \]

where: \( p \) in Pa - pressure; \( \tau \) in Pa - stress tensor; \( g \) in m/s\(^2\) - gravitational acceleration; \( S_i \) - body force (source term).

The source term \( S_i \) of the external body force was modelled using the Darcy Forcheimer for simple homogenous porous medium

\[ S_i = \frac{\Delta p}{\Delta L_i} = -\left(\frac{\mu}{\alpha} v_i + C \frac{1}{2} \rho v_i^2\right) \]

\( \Delta p \) in Pa - pressure drop; \( \mu \) in Pa s - laminar fluid viscosity; \( \alpha \) in m\(^2\) - permeability of the medium; \( C \) in 1/ m - inertial resistance; \( v_i \) in m/s - velocity normal to the porous face in one specific direction; \( \rho \) in kg/m\(^3\) - density; \( \Delta L_i \) in m - thickness of the medium in one specific direction.

Pressure difference is predicted across the domain on inlet fulcrum angle of 40 degrees.

2.1.1. Pre-Processing

2.1.1.1. Meshing

Following steps were taken while meshing of the catalytic converter:

- Trias were introduced where necessary, to maintain the flow and avoid rotating quads.
- Irregularity in element size was avoided as the mesh proceeded.
- Mid surface was used as the design was sheet metal based and to ensure the element deformation is prevented.
- Higher global element size was maintained to reduce processing time.
- Element smoothing was done to approximate the element to ensure a uniform mesh.
- Trias pointing in direction of flow were preferred.
2.1.1.2. Meshing Criteria

- Aspect ratio < 5
- Jacobian ratio > 0.7
- Global element size – 4mm
- Total no. of mesh elements : 460,000
- Minimum quad angle – 45 degrees
- Maximum quad angle – 135 degrees
- Defeaturing < 5mm

2.1.2. Boundary Conditions

Table 2. Properties of the working fluid.

| Working fluid    | Air                  |
|------------------|----------------------|
| Density (kg/m³)  | 0.7534               |
| Operating Temperature | 350°C                |
| Pressure (bar)   | 1.35                 |
| Viscosity (Pa s) | 3.0927*10⁻⁵          |

- Flow Model: k-ε turbulence model
- Boundary condition:
  - Inlet Velocity: 20m/s
  - Operating pressure: 1.35 bar
  - Outlet: pressure outlet
  - Mass flow rate: 43.63kg/hr

2.1.3. CFD Results

The solution converged at 300 iterations for mass flow rate of 0.012 kg/s. Figure 3 and 4 shows the pressure across the catalytic converter at 0.012 kg/s mass flow rate.

![Figure 3. Pressure Drop across CC.](image1)

![Figure 4. Global Pressure Change.](image2)
Figure 5. Outlet zone.

Subsequently Figure 6 and 7 shows the velocity profile. The flow distribution, through the monolith affects the conversion efficiency, catalyst reaction and faster “light-off” during the cold start period. Therefore, design goals include a minimum pressure drop with an ideal flow uniformity index.

Figure 6. Local Velocity Change. 
Figure 7. Velocity Change.

3. Substrate and Catalyst
The substrate is a ceramic structure over which the catalyst is coated. The cell shape of the substrate is hexagonal as suggested most effective in the literature to reduce backpressure and maximum surface area. The substrate is a polyurethane foam with hexagonal cell shape sprayed with alumina silicate powder and was sintered at 600°C. The process was repeated twice in order to ensure a hardened substrate. The substrate was sliced into four equal pieces in order to reduce backpressure and increased strength of the substrate.
For materials, such as ceramics, a sintering mechanism involving the generation of a permanent liquid phase is applied. This type of liquid phase sintering involves the use of an additive to the powder, which will melt before the matrix phase and which will often create a so-called binder phase. At high temperature, the powder melts and moves into the pores via capillary action. Sintering temperature was kept at 500 degree celsius.

The catalyst used is Lanthanum Strontium Manganite which is an oxide ceramic material with a perovskite crystal structure having the general form $\text{ABO}_3$. The crystal consist of A sites occupied by lanthanum atoms and small Manganite atoms on B sites. The lanthanum atoms are doped with strontium atom (+2 oxidation state) which gives it extra holes in valence band. LSM then can reduce the oxygen atoms to oxide atoms which bind with CO and hydrocarbons to give CO$_2$ and water vapour. This material is used to coat cathode in Solid Oxide fuel cells for temperatures upto 600-700 degree Celsius. Lanthanum being a transition metal has catalyzing properties and its doping with strontium helps trap oxygen for oxidation reaction. LSM -20 powder is used having a chemical composition of $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$.

The process used to coat LSM on the substrate is Atmospheric plasma spray which uses the LSM powder being fed into the high temperature ionized gases or plasma that melts the powder particles propelling them onto the surface of the workpiece.

The substrate slices are rolled and packed into the support mat (Ecoflex 400) and was assembled in the shell. The inlet and outlet pipes were made to be removable using bolted flange for good serviceability of the catalyst, mat and inlet pipes.
4. Experimental Setup

The engine used in the testing was Briggs and Stratton BAJA (ATV) 305cc four stroke single cylinder engine and the test was carried out at an Exhaust gas analyser in four different configurations:

- Engine exhaust gas analysed at idling RPM
- Engine exhaust gas analysed at max RPM
- Engine exhaust gas analysed with CC (Heat plug off)
- Engine exhaust gas analysed with CC (Heat plug on)

The catalytic converter was checked for any leaks when fitted with the engine in order to avoid wrong emission results.

The heat plug was given a 12 V supply and was operated by Arduino controlled self-triggered mechanism for 15 seconds. The heat plug was bolted into the inlet pipe near the inlet cone in order to reduce heat loss by the exhaust gases.

The exhaust gas analyser used in testing samples the exhaust gases from the exhaust pipe using probe. It measures the concentration of CO, hydrocarbons, NOx, and oxygen.

The experiment was conducted in the laboratory test facility. Fig.14 shows the catalytic converter bolted with the engine exhaust pipe with heat plug installed. The engine was allowed to run at idling RPM for 5 minutes and readings were noted. Thereafter full throttle readings were taken for next five minutes. Then, engine was allowed to cool for 1 hour and procedure was repeated with Catalytic converter with and without heat plug. Setup gave the emission values to compare with the BS IV emission norms.

![Figure 12. Exhaust Gas Analyser.](image1)

![Figure 13. Water vapour trapped.](image2)
5. Results and emission analysis

Table 3. Emissions results from the Engine.

| Condition                                      | CO % | HC (ppm) | CO2 % | NO (ppm) |
|------------------------------------------------|------|----------|-------|----------|
| Raw emissions-Engine Idling (750)              | 6.2  | 1200     | 5.09  | 81       |
| With the engine at max RPM (3500)              | 5.5  | 1997     | 4.8   | 58       |
| With Catalytic Converter-Idle RPM (750)       | 0.32 | 190      | 9.3   | 56       |
| With CATCON-max RPM (3500 RPM)                | 0.25 | 98       | 10.2  | 72       |
| With CATCON and heat plug on-750rpm           | 0.33 | 188      | 9.1   | 55       |

The results from the experiment shows that the CO and Hydrocarbon emissions are reduced to the level of BS IV emission norms. CO levels as per ARAI BSIV norms is 0.3 vol% at low idle rpm and 0.2 vol% at high rpm for a four wheeler (305cc ATV) petrol engine. The investigation gives a volume percentage 0.32 with catalytic converter at a low idling of 750 RPM and 0.25 vol% at 3500 RPM. The suggested Hydrocarbon levels for a four wheeler petrol engine are 200 ppm which is controlled by the catalytic converter to a level of 188 ppm at 750 RPM and further reduced to 98 ppm at 3500 RPM as the catalyst is fully functioning to carry out reaction effectively as it reaches higher temperature. NO\textsubscript{x} emissions are not reduced as the reduction catalyst was not used but LSM if coupled with Rhodium (reduction catalyst to reduce No\textsubscript{x} emissions) can prove to be an effective emission reduction setup.
Also, the time taken by the catalyst to start the oxidizing reaction was found to be 50 seconds, evidenced by no reduction in HC emission levels even with the catalytic converter. But as the heat plug is switched on for 15 seconds, the time taken was 35 seconds which shows the emission levels produced in 15 seconds were reduced. Hence, pre-heating technique has proved to be useful to some extent even by using an ordinary heat plug used in automobiles.

The NOx emissions were reduced after some time which is a possible indication of high backpressure due to the increased exhaust gases remaining in the cylinder. The emission levels were reduced for 45 minutes which included both 30 minutes of engine running at idling RPM of 750 and 15 minutes full throttle RPM of 3500. But then CO emissions were increased along with decreased oxygen content. This shows that the LSM catalyst might have degraded and was not able to trap the oxygen sufficiently after 45 minutes.

6. Conclusions and future research
This research included design, analysis and fabrication of a catalytic converter with a new LSM catalyst primarily for oxidation of Carbon Monoxide and Hydrocarbons. So, following are the main conclusions from the performed experimental and numerical study:

- Lanthanum Strontium Manganite is a prospective candidate for the oxidation catalyst in catalytic converters as it can reduce CO and HC emissions.
- LSM in different compositions can help reach the emissions to the level of BS VI norms and its catalytic properties deteriorate after a period of 45 minutes of engine running continuously with the catalytic converter.
- The cost of the catalyst of 10 gms LSM 20 was found to be Rs. 4450 which was coated over the substrate. This cost is almost 10 times lesser of Platinum which costs Rs. 45,000 in Indian market.
- NOx emissions reduced after a period of time which is an evidence of increased backpressure which is possibly due to the sliced substrates.

Current study has the following scope for further research:
- Temperature sensor supported Heat Plug.
- Different compositions of Lanthanum and strontium in perovskite.
- Ultrasonic treatment for catalyst to improve the life of catalyst.

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