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Low Cost Catalytic Converter Retrofit using Ceria and Zirconia Coated Monoliths for Old Diesel Automobiles to control pollution

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Abstract:

Automobile pollution plays a key role on the impact of the health hazard to living being. Emissions from Diesel vehicles are more hazardous because of soot besides the harmful pollutants like CO, HC, CO_2 and NOx. As the BS VI pollution norms are in force and many vehicles are left unsold and lying at the premises of the car factories and its becoming a huge loss to them. The authors of this paper tried to reduce the level of pollutants leaving to the atmosphere from the diesel engines by developing a retrofit catalytic converter and also using the Palash Bio diesel blends. The authors have tried different materials for the monoliths like Aluminum, Copper, SS and Pot makers' Clay with different catalyst coating materials like CeO_2 (Ceria), ZrO_2(Zirconia) and TiO_2 which are less expensive and hence makes the retrofit catalytic converter affordable by the middle class people also so as to make their cars compatible to BS VI norms. The authors have obtained an Indian Patent also in this regard. The experimental results have shown significant reduction in pollutants to the extent of 55 to 65% reduction with the use of Retrofit Catalytic Converter and further reduced by using the Palashbio diesel blends.

Key words: Retrofit, Catalytic Converter, Bio Diesel, Ceria, Zirconia, Pollution Control

Introduction:

Automotive vehicles emit several exhaust gases and pollutants. Diesel vehicles are more polluting because of emission of soot, which is particulate matter. The exhaust gases contain N_2, H_2O and CO_2 which are not toxic. Whereas harmful gases like CO are produced due to incomplete combustion of fuel and NOx is produced at high temperatures. The Euro 6 norms have become very stringent because of NOx and PM are drastically reduced. (Table 1a) The Indian government has adapted the Euro norms as BS IV and decided to leap frog to BS VI by 2020. This decision has put lot of pressure on auto Industry. [2] Till April 2017, BS IV norms were made applicable in 13 major cities while BS III norms were applicable in other areas of India. However in January 2016 the Central Govt. of India decided to skip BS V norms and replaced it with the new BS VI norms fromApril 2020. [3]

Table 1a: Emission Standards of India for CIengine vehicles (Light duty) [3]

Diesel Vehicles	Unit	BS IV Norms M and N1 Cls I	BS IV Norms N1 Cls II	BS IV Norms N1 Cls III	BS VI Norms M and N1 Cls I	BS VI Norms N1 Cls II	BS VI Norms N1 Cls III
NOx	**g/km**	0.25	0.33	0.39	0.08	0.105	0.125
PM	**g/km**	0.025	0.04	0.06	0.0045	0.0045	0.0045
HC+NOx	**g/km**	0.3	0.39	0.46	0.17	0.195	0.215
CO	**g/km**	0.5	0.63	0.74	0.5	0.63	0.74

- M category include motor vehicles having at least 4 wheels and for passenger carriage.
- N1 Class I include vehicles having at least 4 wheels used for goods carriage <3.5 tonnes
- N1 Class II same as above but for loads>3.5<12tonnes
- N1 Class III Same as above but for loads>12 tons

Source: http://transportpolicy.net/index.php?title=India:_Light_duty:_emissions.

Table 1 b: On Board Diagnostics Threshold values of Compression Ignition Engine. [3]

Norm	OBD Threshold Limits for Compression Ignition Engine, g/km					
	Dt.	**Vehicle category**	**CO**	**NMHC**	**NO_x**	**PM**
BS VI–1	**1st Apr 2020**	M &N1 cls I	1.75	0.29	0.18	0.025
		N1 cls II	2.2	0.32	0.22	0.025
		N1 cls III	2.5	0.35	0.28	0.03
BS VI–2	**1st Apr 2023**	M &N1 cls I	1.75	0.29	0.14	0.012
		N1 cls II	2.2	0.32	0.18	0.012
		N1 cls III	2.5	0.35	0.22	0.012

The implementation of the new BS VI emission norms need to reduce nitrogen oxides (NOx) and particulate matter (PM) by around 68% and 80% respectively in diesel vehicles.Significant engine technology changes including increased injection & cylinder pressures, improvements in engine combustion & calibration and exhaust emission after treatment are required to be changed to a great extent for the implementation of these new emission norms, and thus putting immense pressure on auto Industry.

In case of CI engines three devices namely Catalytic converter, Diesel particulate filter and Selective catalytic reduction are fitted in series, to make them BS VI compliant. Thus evolution offour-way catalytic converter took place, which removes all four major pollutants namely HC (unburnt hydrocarbons), PM (Particulate matter), CO & NOx.

On Road Real Driving Emissions:

The European Union (EU) wanted a white paper on pollution control as the pollutants are likely to reach alarming levels by 2025. ICCT have given a white paper in 2015.[1] This has special reference to failure of Euronorms in real worlddriving. Usually pollution certificates are issued based on a test conducted on a stationery car with engine idling condition. It has been found that in the real world driving the CO and NOx (major pollutants) levels are very high due to slow driving and traffic conditions & congestion at traffic lights. They suggested checking Real Driving Emissions (RDE) should be done and record the pollution levels in the onboard instruments in the carduring driving under different conditions. They found that NOx levels reach 300 to 400 mg/km incomparison to 80 mg/km stipulated in Euro VI norms. Presently a confirmatory factor of 5.7 is used to equate Euro VI to RDE Levels. The report suggest that this factor should be cut down to 2.1 by 2017 and then to 1.5 by end of 2020.

India has adopted the OBD-1 and OBD-2 from years 2010 and 2013 respectively. Since 1st April 2020 onwards the BS VI-1 OBD is being implemented in all the vehicles. Its been proposed that from 1st April 2023 the BS VI-2 OBD is to be implemented and the threshold values of the OBD are given in table 1b.

Methodology:

The authors of this paper have taken up design and development of an automobile catalytic converter which will make old diesel vehicles compatible with BS VI pollution norms. The Indian govt. has finalized the vehicle scraping policy and made legislation. As per this all vehicles above age of 15 years should be scrapped. The transport sector which uses vans, trucks & buses which are more than 15 years old and as per new legislation all these entrepreneurs will be out of business.

India as it is a developing country may not afford to literally scrap all old vehicles like in USA and Europe. By using suitable extra fittings like EGR, SCR, DPF and catalytic converter these vehicles can be given a new lease of life to make them compatible with BS VI norms which are in force from April 2020.

The authors are trying to contribute their mite to solve the problem of BS VI compatibility of automobiles. At the same time it has to be considered that those people running cabs, trucks & buses cannot afford to add EGR, SCR as extra equipment. By using a cost effective 4 way catalytic converter retrofit, they can overcome the BS VI threat.

Design of Four Way Catalytic Converter

The catalytic converter has the following components:

1. Inlet diffuser
2. Oxidation Monolith
3. Reduction Monolith
4. Outlet Reducer.

The inlet and outlet diffuser & reducer angles are optimized based on pressure drop,Δp. (fig 6)

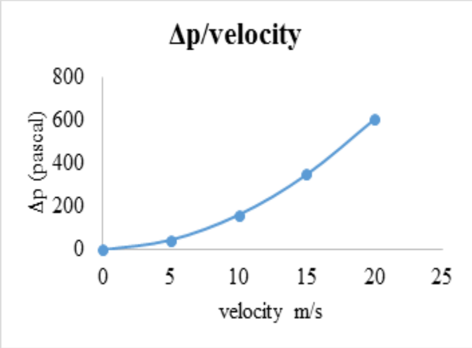

Fig 6: Pressure Vs Velocity

Polynomial is assumed in the process and the equation based on the curve is:

$Y = 1.4x^2+2.189x+2E\text{-}1.3$
Polynomial equation for unit length is
$\Delta p = 1.4V^2 +2.189\ V$
Using this polynomial the pressure drop through Catalytic converter is as follows:

Table 2: Velocity vs Pressure drop

Velocity, m/s	Pressure drop Δp (Pascal)
0	0
5	45.9
10	161.89
15	347.84
20	603.78

Volume flow rate of exhaust gas = Swept Volume x No. of intake strokes per hour

(Assuming dia of cylinder = 87.5mm; Single Cylinder, stroke = 110 mm; 4 stroke Laboratory diesel engine)

Volume flow rate = 29.31 m^3/hr

Exhaust pipe dia = 2"; Area of X^n = 0.0078 m^2
Velocity = 5 m/s to 6m/s.

1. Optimization of inlet diffuser and outlet Reducer angles:

Using Computational fluid dynamics software the diffuser/reducer flare angles optimized by considering minimum pressure drop. The results of which are given in table 3 a & b.

Table 3 (a): Pressure drop values in catalytic converter with varying inlet angle and constant outlet angle.

Velocity (m/s)	**Pressure drop (Pascal)**					
	15^0-45^0angles	**25^0-45^0 angles**	**35^0-45^0 Angles**	**45^0-45^0 angles**	**55^0-45^0 angles**	**65^0-45^0angles**
5	300	300	302	303	306	310
15	2699	2703	2714	2733	2759	2803
25	7495	7506	7546	7603	7677	7808

Table 3 (b): Pressure drop values in catalytic converter for same inlet and outlet angles.

Velocity (m/s)	**Pressure drop (Pascal)**				
	15^0-15^0 angles	**25^0-25^0 angles**	**35^0-35^0 angles**	**45^0-45^0 angles**	**65^0-65^0 angles**
5	299	300	301	303	316
15	2688	2693	2702	2733	2869
25	7467	7480	7506	7603	8040

It has been decided based on the above data to have a 25^0 for both inlet and outlet for minimum pressure drop even though 15^0 is giving lesser pressure drop for the ease of fabrication of the housing the 25^0 has been chosen as shown in fig 7.

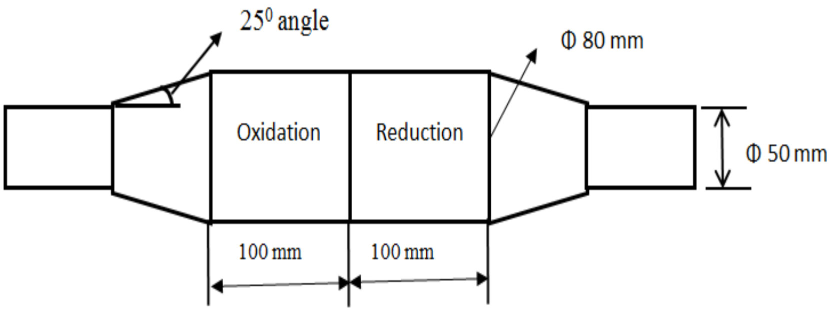

Fig 7: Final catalytic converter housing with optimized angles.

2. Optimization of Monolith Channel cross section.

Different cross sections have been examined usingCFD Software - Square, Triangle, Circle and Hexagonal. The optimized cross section appeared as hexagonal. See table 4.

Table 4: Pressure drop values for different shapes of channel openings

Velocity (m/s)	Pressure drop (Pascal)			
	hexagon	Circular	Square	Triangular
5	48	47	54	69
5.8	64	64	73	93
10	186	186	210	259
15	401	406	451	541
20	686	704	768	903

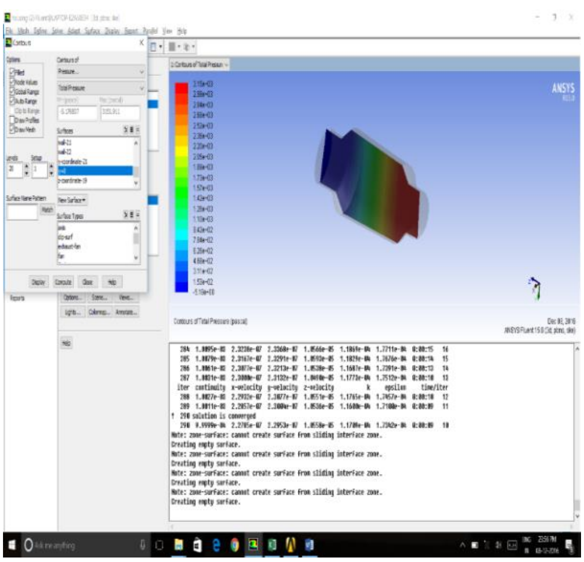

Fig8a: Square:

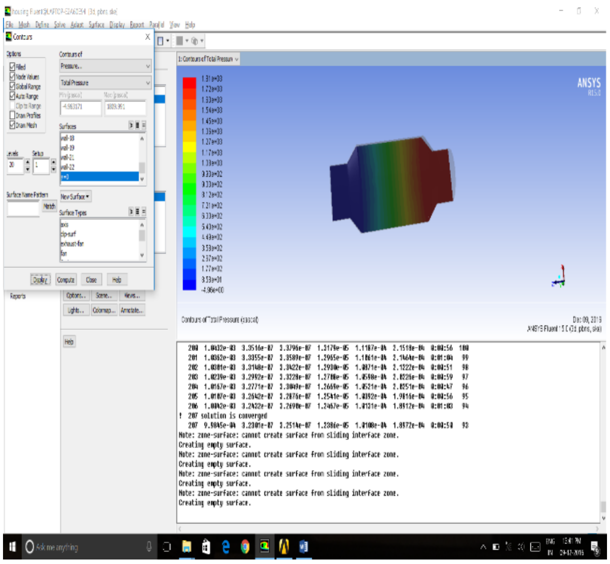

Fig 8b: Triangle:

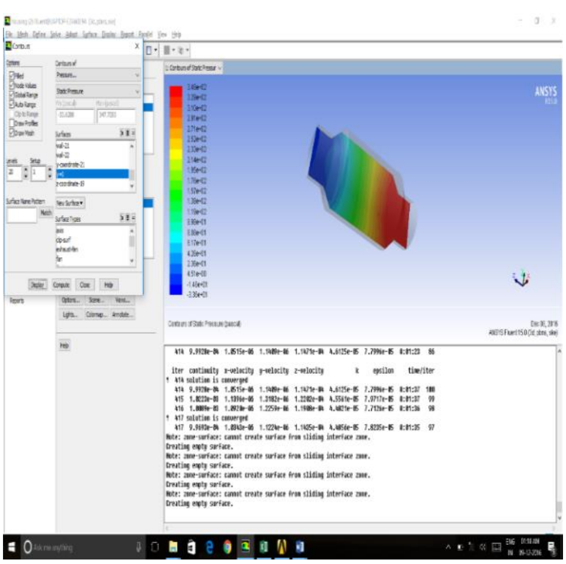

Fig 8c: Circle

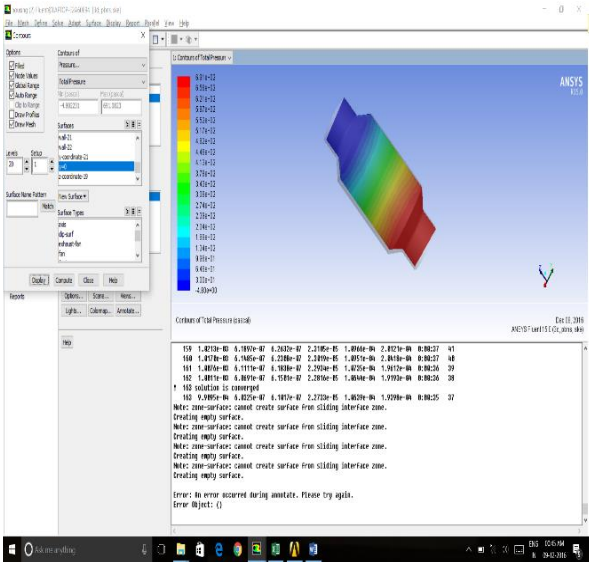

Fig 8d:Hexagon

Fig 8: Total Pressure drop across different channels

Design of Monolith shell dimensions

Volume flow rate of exhaust gas = Swept Volume x Number of intake strokes/hr

$$= \frac{\pi}{4} D^2 L \times \frac{rpm}{2} \times 60$$

Catalyst volume = Volume flow rate / Space Velocity

Space Velocity = 6 m/sx3600 = 20,000/hr.

$$V_{catalyst} = \frac{\pi}{4} D^2 L$$

assumeL=2D

Calculate D, and L

Typicalcalculation for Chemical Reactions:

Conversion of pollutants CO and C_6H_6 to CO_2

$2CO + O_2 \longrightarrow 2CO_2$ ---- (eq. 1)

$2C_6H_6 + 15O_2 \longrightarrow 12\ CO_2 + 6\ H_2O$ ---- (eq. 2)

CO converted $\times \frac{(mole*molecular\ wt\ CO2)}{(mole*molecular\ wt\ CO)}$

η = conversion efficiency

$= 40.2$ g/mile CO $\times \frac{2x44.009\ of\ CO2}{2x28.01\ of\ CO}$

C_6H_6 converted $* \frac{(mole*molecular\ wt\ CO2)}{(mole*molecular\ wt\ C6H6)}$

$= 40.18$ g/mile $C_6H_6 \frac{12x44.0009\ of\ CO2}{2x78.114\ of\ C6H6}$

$= 3.38$ g/mile CO_2.

Here the values of CO converted and C_6H_6 converted (HC) are taken from experimental values.

CO Conversion efficiency = 100 x (CO_{inlet} - CO_{outlet}) / CO_{inlet}

HC Conversion efficiency = 100 x ($C_6H_{6\ inlet}$ – $C_6H_{6\ outlet}$) / $C_6H_{6\ inlet}$

(Here C_6H_6 is unburnt HC in exhaust gas)

The final prototype catalytic converter is shown in fig 9.

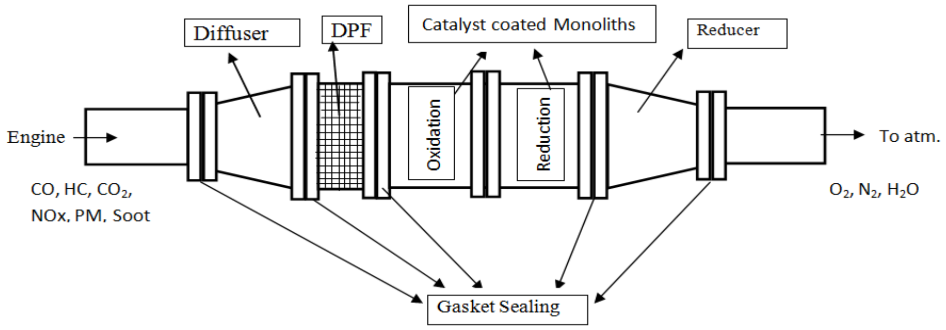

Fig 9: Four way Catalytic converter

4.0 Experimentation

Optimization of Catalytic Converter Retrofit:

Basically a 4 way catalytic converter consists of Diffuser, Oxidation Monolith, Reduction monolith and Reducer along with a diesel particulate filter as shown in Fig 8. The monolith is made of metal or ceramic material on which the catalysts (Oxidation/Reduction) are coated. In the original equipment of Catalytic converter, the monolith used to be of ceramic material and coated with noble metals like Pt, Pd or Rh or combination. However noble metals are expensive and becoming scarce.

Hence researchers all over the world concentrated on finding substitute catalysts for noble metals. Chemical compounds like Cerium oxide, Zirconium Oxide, Titanium dioxide appear to cater to the requirement of both oxidation and reduction catalysts. Also ceramic monolith, though highly heat resistant, are heavy and expensive to manufacture. Hence metal monoliths were tried. Metals like Copper, Aluminium, Steel and Nickel were found to be good catalyst supporters. Using the expertise available in the published literature, the authors fabricated metal monoliths of various types like Copper, Aluminium& Steel with corrugated sheets or discs with holes.

Also the authors found that pot makers' clay works as a good Catalyst support. Marbles of different sizes were prepared from the pot makers' clay and tried and found 10mm dia marbles are the best. An Indian patent was granted to the authors with Patent No: 365869. [18]

Finally authors of this paper zeroed in on eight monolith models and conducted experiments and selected the following top four performing monoliths which arecoated with suitable coatings like CeO_2, ZrO_2, TiO_2 with different combinations: [16, 17]

1. 10mm clay marbles coated with CeO_2, ZrO_2 enclosed in perforated steel canisters
2. Corrugated Aluminium sheet coated with CeO_2, ZrO_2
3. Corrugated Aluminium sheet coated with TiO_2
4. Corrugated Copper sheet coated with CeO_2&ZrO_2

In all the above retrofit models Alumina (Al_2O_3) wash coat was used before coating with catalysts. Out of the 8 models, the top four were compared and given in table 5.

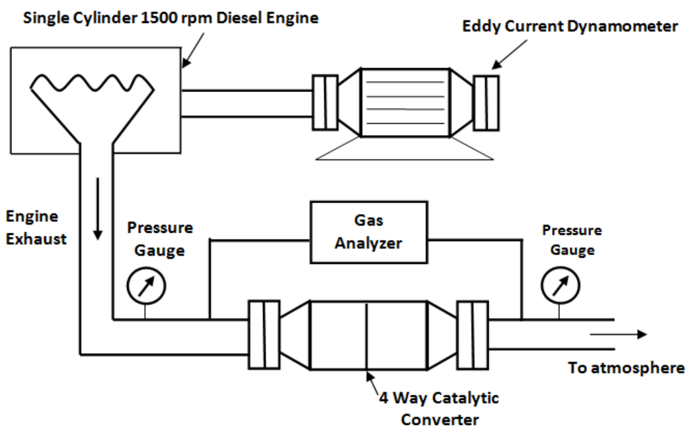

Fig 10: Schematic diagram of test set up in Laboratory

The laboratory test set up is shown in Fig 10. The laboratory experimental results of top four models as given in Table 5 show that the 10mm clay marbles and aluminium metal sheets were promising monolith supports. A retrofit with 10mm clay marbles coated with Aluminium Oxide as wash coat and CeO_2&ZrO_2 as Catalysts was identified for the purpose of reduction in CO, CO_2, HC & NOx levels. Moreover this is the cheapest of all the models. Hence if a cost effective model is to be brought out commercially, 10mm dia clay marble monoliths coated with CeO_2&ZrO_2 can be chosen.

Table 5: Conversion efficiencies of Top 4 performing monoliths with diesel as fuel

S.No	Monolith Material & Coatings	Load, Kg	Conversion Efficiency %				
			CO	CO_2	HC	O_2	NOx
1	Corrugated Aluminum with CeO_2& ZrO_2	2	53.13	43.15	57.97	29.00	75.76
		4	54.15	49.39	54.75	41.52	66.27
		6	52.68	49.72	52.86	55.53	72.27
		8	61.66	55.81	52.91	109.23	64.39
		10	65.02	57.19	52.38	136.59	62.11
2	Corrugated Aluminum with TiO_2& TiO_2	2	50.00	39.26	59.42	28.27	83.33
		4	50.87	47.42	53.48	38.83	77.11
		6	48.07	48.60	53.43	54.31	76.47
		8	57.67	54.37	50.24	110.21	68.18
		10	63.49	56.32	48.53	140.65	57.76
3	Copper Corrugated with CeO_2& ZrO_2	2	41.18	27.22	52.54	19.12	68.18
		4	37.88	35.61	47.47	26.01	67.47
		6	34.01	33.99	42.29	38.38	67.23
		8	42.81	43.35	37.38	89.09	59.85
		10	46.77	48.43	36.08	116.91	51.55
4	10 mm Clay Marbles with CeO_2 and ZrO_2	2	37.12	21.30	58.33	14.22	89.39
		4	33.19	26.97	45.25	22.15	80.72
		6	25.54	27.81	39.71	31.27	75.63
		8	41.21	31.98	37.86	69.51	65.15
		10	43.10	34.38	34.80	86.18	50.93

5.0 Bio diesel Blends as fuel:

Indian govt. has envisaged blending of bio diesel with diesel and started experimentally in national capital with 10% blend. The main reason in that the govt. wants to reduce volume of crude import from gulf countries.

Uma MaheswaraRao, H Suresh BabuRao, MalothRamuluNaik and others [13, 14, 15] have studied the bio diesel blends performance in diesel engines. Variable Compression Ratio (VCR) diesel engine gives a lot of flexibility in handling bio diesel blend as fuel, as it causes additional smoke, which can be reduced by VCR. All these published literature have reiterated that the use of bio diesel blend reduces exhaust pollution levels compared to pure diesel fuel.

The authors conducted laboratory tests (see Fig. 10) on this aspect with Palash bio Diesel with B20 blend and the results given in tables 6, 7, 8 and 9 indicate the level of advantage of using bio diesel blends.

Table 6. Results of Diesel engine exhaust emissions without catalytic converter with diesel fuel.

Load, Kg	Without Catalytic Converter				
	CO % vol	CO_2 % vol	HC ppm	O_2 % vol	NOx ppm
2	0.86	5.4	276	12.45	66
4	0.91	6.6	316	11.15	83
6	0.93	7.12	350	9.85	119
8	1.25	8.35	412	7.15	132
10	1.5	9.25	546	6.15	161

Table 7. Results of Diesel engine exhaust emissions without catalytic converter with B20 Palashbio diesel and diesel mix.

Load, Kg	Without Catalytic Converter				
	CO % vol	CO_2 % vol	HC ppm	O_2 % vol	NOx ppm
2	0.72	5.2	142	12	44
4	0.77	6.1	212	10.9	69
6	0.81	7	297	9.5	85
8	0.95	7.9	375	6.85	148
10	1.15	9	490	6	170

B20 fuel: 80% diesel oil + 20% Palash biodiesel oil

Table 8.Results of diesel engine emissions with Catalytic converter with 10 mm Clay marbles monolith with diesel fuel.

Load, Kg	With Catalytic Converter				
	CO % vol	CO_2 % vol	HC ppm	O_2 % vol	NOx ppm
2	0.54	4.25	115	14.22	7
4	0.61	4.82	173	13.62	16
6	0.69	5.14	211	12.93	29
8	0.73	5.68	256	12.12	46
10	0.85	6.07	356	11.45	79

Table 9.Results of diesel engine emissionswith catalytic converter with 10 mm Clay marbles monolith with B20 fuel.

Load, Kg	With Catalytic Converter				
	CO % vol	CO_2 % vol	HC ppm	O_2 % vol	NOx ppm
2	0.48	4.11	59	13.4	4
4	0.51	4.5	96	12.73	9
6	0.54	5.11	133	12.05	21
8	0.68	5.35	154	11.5	54
10	0.79	5.98	196	10.7	83

The emission levels of Bio diesel and Diesel without the Catalytic Converter are represented graphically in Fig 11 & 12.

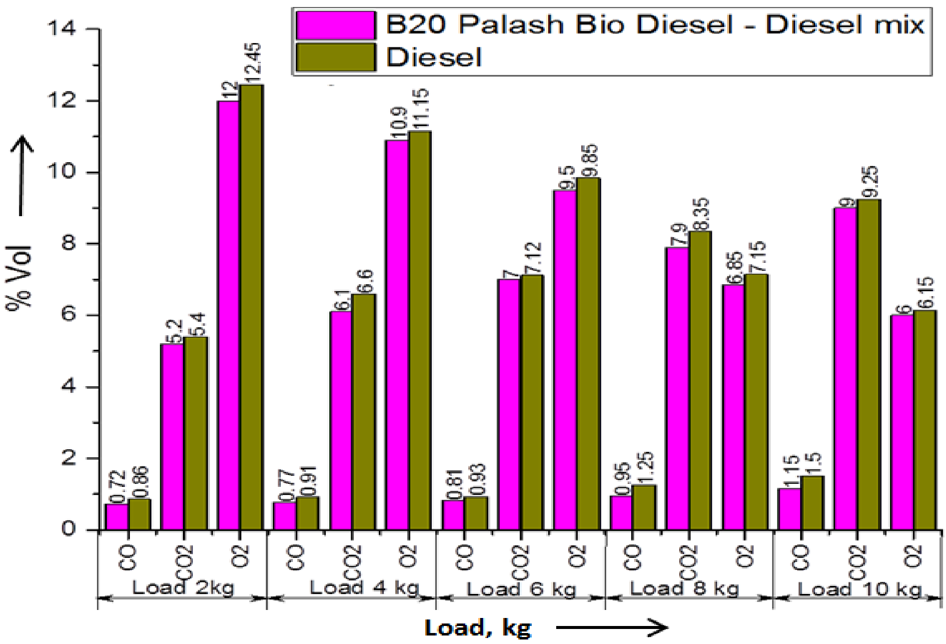

Fig 11: Comparison of CO, CO_2& O_2 emissions between Bio Diesel and Diesel without catalytic converter

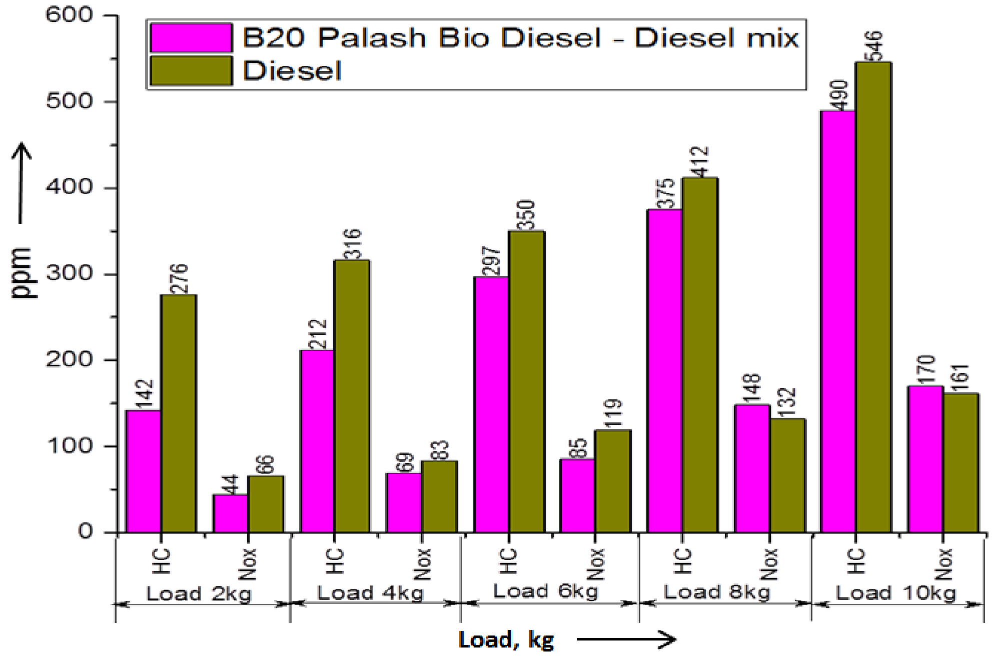

Fig 12: Comparison of HC&NOx emissions between Bio Diesel and Diesel without catalytic converter

The emission levels of Bio diesel and Diesel when fitted with the Catalytic Converter with clay marbles monolith are represented graphically in Fig 13& 14.

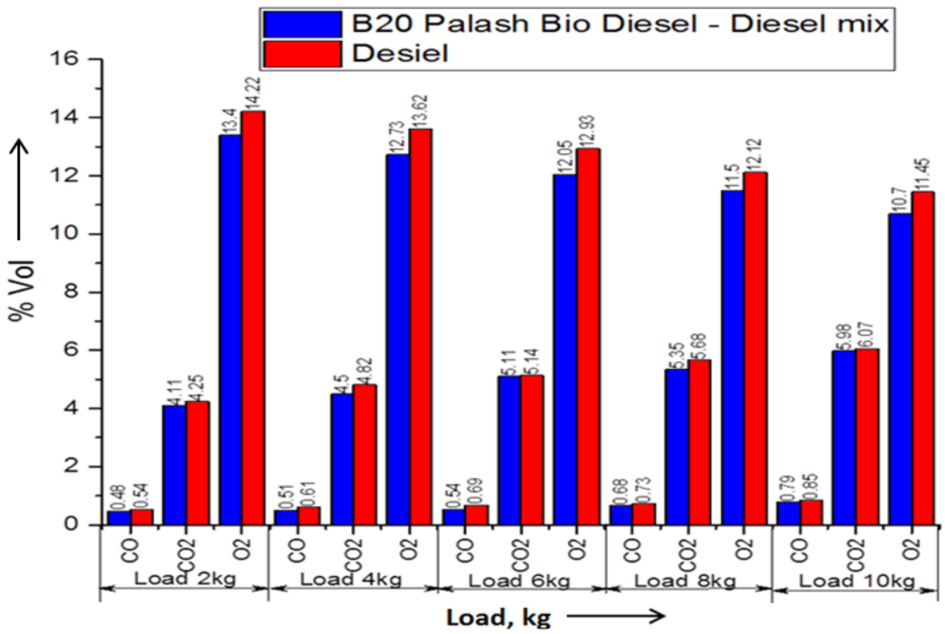

Fig 13: Comparison of CO, CO_2& O_2 emissions between Bio Diesel and Diesel with catalytic converter

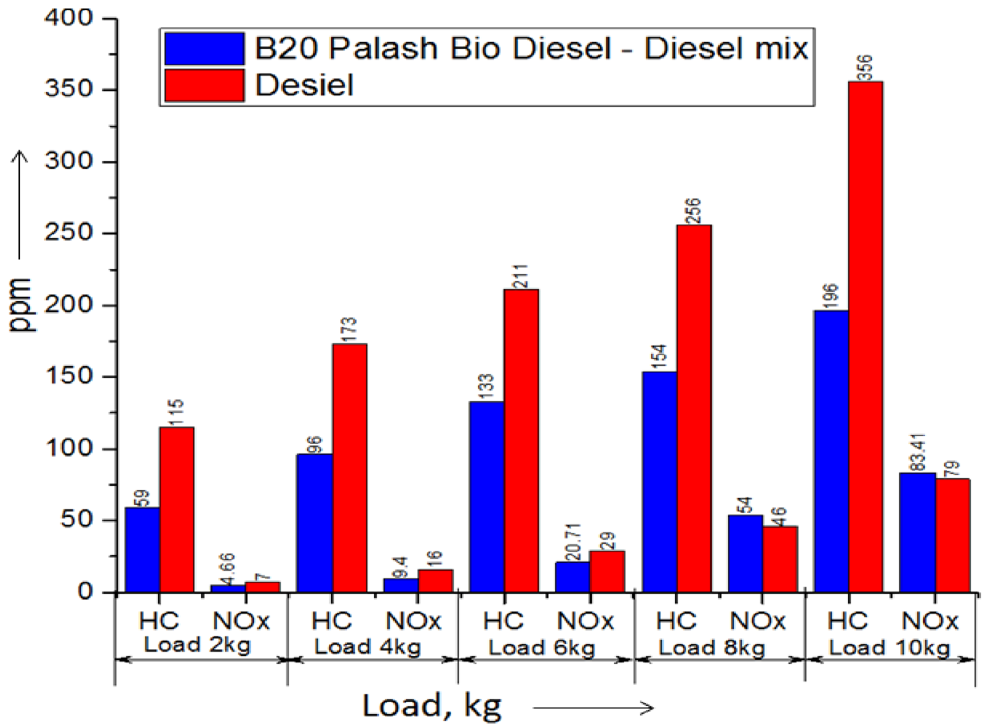

Fig 14: Comparison of HC&NOx emissions between Bio Diesel and Diesel with catalytic converter

6.0 Discussion of Results:

1. Using computational fluid dynamics software - fluent, the authors optimized flow channels and selected suitable flow channel which gives minimum back pressure. [16]

2. In the original catalytic converter provided as equipment with high value cars like BMW, Benz etc., Pt, Pd and Rh noble metals are used as catalyst coatings on the ceramic monoliths. The authors experimented with Cu, Al and SS metal monoliths and Pot makers' clay marbles as monolith materials and coated with CeO_2, ZrO_2 and TiO_2 as catalyst coatings over Al_2O_3 wash coat. By conducting laboratory experiments it was optimized by performance based on the conversion efficiencies, zeroed in on 10 mm clay marbles kept in canisters as monolith.

3. The authors tried Palash oil bio diesel blended with diesel. B20 blend, which corresponds to 20% bio diesel & 80% diesel appeared suitable and reduced pollution levels further.

4. The difference in reduction of pollutants when the Catalytic Converter fitted to the exhaust pipe for both B20 Bio diesel blend and pure Diesel used as fuels is shown in table 10.

5. It is evident from Table 10 that the pollutants like CO, CO_2, HC & NOx have been reduced by 37 to 43% CO, 21 to 34% CO2, 58 to 34% HC & 89 to 50% NOx at 2 kg to 6 kg loads respectively for the Diesel fuel and further reduced by 30% CO, 20 to 33% CO2, 54 to 60% HC and 90 to 51% NOx respectively at 2 kg to 6 kg loads for B20 Palash Bio Diesel blend fuel when the Catalytic Converter with Clay marbles coated with CeO_2&ZrO_2 as monoliths is fitted to the exhaust pipe.

6. As a diesel particulate filter is incorporated in the Catalytic Converter the soot and particulate matter emission is drastically reduced.

7. All the pollutants have been significantly reduced when the B20 Palash Bio diesel blend is used as fuel rather than the diesel and further reduced with the use of the Catalytic Converter.

8. The percentage of reduction is more for the medium loads i.e 4 & 6 kgswhereas at higher loads it is less.

9. The NOx emissions have been increased for higher loads whenB20 Bio diesel blend is used which is inevitable for any bio diesel and the amount of increase of NOx emission is in between 9 to 16 ppm at 8 and 10 kg loads and its percentage of increase is around 5.5 to 17 %.

10. By the use of the Catalytic converter with Clay marbles as monoliths the increase in NOx emission at higher loads has been decreased to

Table 10: Reduction of pollutants for B20 Bio diesel blend &Diesel with Catalytic Converter

S.No	Type of Catalytic Converter	Load, kg	Difference in reduction of pollutants				
			CO %	CO_2 %	HC %	O_2 %	NOx %
1	B20 Palash Bio Diesel - Diesel mix	2	33.33	20.96	58.45	11.67	90.91
		4	33.77	26.23	54.72	16.79	86.96
		6	33.33	27.00	55.22	26.84	75.29
		8	28.42	32.28	58.93	67.88	63.51
		10	31.30	33.56	60.00	78.33	51.18
2	Diesel	2	37.21	21.30	58.33	14.22	89.39
		4	32.97	26.97	45.25	22.15	80.72
		6	25.81	27.81	39.71	31.27	75.63
		8	41.60	31.98	37.86	69.51	65.15
		10	43.33	34.38	34.80	86.18	50.93

7.0 Conclusions:

The authors of this paper have successfully developed a retrofit of catalytic converter which is cost effective and can be used as extra equipment to make old diesel automobiles BS VI compatible. Also using bio diesel blends give extra leverage to reduce pollution levels to be on par with stipulated BS VI norms.

BS VI norms stipulate PM level of 0.0045 gm/km and NOx value of 0.06 to 0.085 gm/km. The final values obtained with retrofit developed are HC 59 ppm and NOx 4.66 ppm at 2 kg load on the engine. It may be noted that the pollution levels are measured in engine idle conditions and 2 kg load on dynamometer corresponds to the PUC (Pollution Under Control) test conditions.

However, the authors reiterate that as the results are highly encouraging, by fine tuning laboratory test conditions the results can be far better than shown in this paper. The authors got a patent for the catalytic converter retrofit. [18].

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