SAE International

2012-01-1672
Published 09/10/2012
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doi:10.4271/2012-01-1672
saeeng.saejournals.org

Mileage Influence on Conversion Efficiency of Catalytic Converter from In-Use Vehicles

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ABSTRACT

Although many works are published about the achieved advancements in the manufacturing of the catalytic converters (CC) system for vehicle engines and their testing under laboratory conditions, there is a lack in the published research about the mileages influence on their conversion efficiency (CE). Dependence of dual-brick CCs' CE in real-world driving conditions on vehicle mileage is studied for the first time. The CC tested are dismantled from the vehicles with mileage from 0 (new one) up to 150000 km. The studied CC are evaluated at the engine test bench containing a dynamometer coupled with a spark ignition engine suitable for this type of CC system. Measurements of CC efficiency are performed at four different engine operation regimes: two loaded regimes and two non-load regimes - low and high speed idling. It is found that the oxidation of CO and HC at all four tested regimes took place almost totally in the first CC. Under higher engine loads the conversion of NO_x also took place mainly in the first CC. The overall efficiency of conversion reaches values above 99% for most of the tested systems. No significant degradation of CE is found in the studied vehicle mileage range. CE of the CC system that has accumulated the highest mileage, of almost 150000 km, reaches values of 97-99% for CO, 86-99% for HC and 64-99% for NOx at the loaded testing regimes. The smaller values are obtained for the low load regime when the CC temperature is decreased. Considering that the European legislation imposes a durability of 160000 km for the CCs, the results obtained in this work show that for the mileage of 150000 km CC keep a very high CE rate. The CC system that included an aftermarket non-genuine CC is the only one, where significant catalyst deactivation is observed. Registered values of the backpressure do not show any of the tested CC systems as being clogged.

CITATION: Tartakovsky, L., Baibikov, V., Veinblat, M., Popescu, D. et al., "Mileage Influence on Conversion Efficiency of Catalytic Converter from In-Use Vehicles," *SAE Int. J. Engines* 5(4):2012, doi:10.4271/2012-01-1672.

INTRODUCTION

In order to meet severe emission standards for gasoline fueled vehicles with spark ignition (SI) engines, thermally durable three-way catalyst (TWC) with enhanced activity has been developed. To achieve the goal of development of highly efficient TWC many efforts were spent and described in the numerous publications.

The implementation of the new regulations concerning the emission levels require fast catalyst light off and fast closed-loop control through high speed engine management. For this purpose the novel fast light off catalyst with perforated foil technology was developed, which showed superior emission light-off characteristic and robustness [1]. To satisfy the requirements concerning the durability at high exhaust gas temperatures (above 1000°C), an advanced metal substrate using newly developed stainless Fe-Cr-Al foil with more than 7.5 mass per cent of aluminum was created [2]. Substrates featuring high cell density combined with ultra thin walls were developed. They offer significant improvement in catalytic efficiency, because of the increased geometric surface area and the reduced thermal mass [3, 4].

At the same time, the position of the catalytic converter has been moved closer to the engine [4]. To obtain high flexibility in converter design, various technologies were proposed [5, 6], e.g. modular block metallic substrate. The latter consists of pre-coated metallic foils, which are welded to a metallic frame and form a substrate block [6]. Another direction of TWC improvement was the increase in usage of palladium as catalyst [7,8,9]. Williamson et al.[8] show that incorporating the Pd-only technologies into close-coupled plus under floor systems, dual brick close-coupled converters, or into close-coupled Pd/Rh systems will be the more likely solutions for achieving optimal emissions, especially for high speed / hard accelerations as occur in the European EUDC or US06 driving cycle. Results of the study [9] indicate that dual brick catalyst systems containing Pd-only catalysts followed by Pt/Rh three-way catalysts provide an effective strategy for managing Pt, Pd and Rh precious metal inventories while achieving LEV / ULEV emission standards.

However, the higher thermal loads exerted on CC system is the disadvantage arising from its placement near the engine. As a result of consistent further development of new washcoat formulations, temperatures of 1050°C now can be attained without impairment of the coating. The high temperature coating is capable of withstanding temperature up to 1050°C for 24 hours and still maintains very high conversion efficiency, thus allowing vehicle manufacturers to install the CC system closer to engine [10]. The idea of using thermally stable CeO_2 / ZrO_2 oxygen storage washcoat materials was published [11, 12]. In addition to their thermal stability, increase of the bulk oxygen mobility of the CeO_2 / ZrO_2 mixed oxides is of great interest to boost the conversion of the pollutants under transient modes and during the cold operations as well. In the forthcoming years advances are expected in nanofabrication techniques that should enable precise control over the shape, size and location of the catalyst to increase the reaction speed, yield and catalyst durability [13].

Although significant progress was achieved recently in the development of new TWC, their efficiency is reduced with aging. Therefore, comprehensive study is required on automotive catalysts deactivation as a function of vehicle mileage. It has been shown that there are many independent processes that collectively contribute to catalyst deactivation [14]. The most important of them are chemical and thermal mechanisms [15, 16]. Chemical deactivation mechanisms primarily consist of constituent poisoning and surface glazing (Zn, Ca and / or P containing phases). Thermal deactivation results in micro structural changes in the washcoat constituents and the precious metals. These thermally induced micro structural effects include sintering, alloying, particle size changes, phase transformations, and constituent interactions.

Arapatsakos and Sparis [17] showed that the CE of a TWC gradually deteriorates owing to thermal, chemical and mechanical effects. If the case of abrupt destruction as a result of extreme mechanical or thermal stresses is excluded, it is generally accepted that, under normal operating conditions, the deterioration in catalyst efficiency due to chemical deactivation originates from the entrance sections of the converter and gradually progresses towards the exit. Some of the chemical effects that cause catalyst efficiency reduction are reversible, such as HC and CO storage due to a temporary λ sensor malfunction or engine misfire. Other processes are considered to be more permanent, such as lead, sulfur and Zn poisoning. Thermal effects such Pt/Rh and Pd/Rh sintering are irreversible. Ball et al. [18] showed that phosphorus originating from the engine oil poisons the surface of automotive catalyst. In general it is believed that as engine oil is consumed, zinc-phosphorus compounds form a diffusion barrier on the surface of the catalyst affecting its performance. It was found that non-combusted zinc-dialkyl-dithiophosphate (ZDP) in the exhaust can form a zinc-phosphate glaze over the catalyst, thus significantly reducing its activity. It is known that ZDP has been used for decades as an effective anti-wear, anti-oxidant ingredient in automotive engine oils. Due to the mentioned above reasons, in the last years the content of ZDP in engine oil has been reduced. Beck et al. [19] noted that calcium and magnesium sulfonates may also be present in the engine oil as detergents and their impact on catalyst performance has not been clearly identified. Some investigators have claimed a net advantage of these agents in preventing the deposition of phosphorous and zinc compounds [19].

Although many works were published about the continuing improvements of CC systems for SI engines and about the testing of those systems under laboratory conditions to estimate the CE as a function of the aging, no information is available about a dependence of CC conversion efficiency in real-world driving conditions on vehicle mileage. The aim of this study was to assess experimentally the CE of in-use CC system as a function of vehicle mileage and to compare these results with the CE of a new CC.

METHODOLOGY

For the purpose of this study, number of dual-brick CC systems have been dismantled from in-use passenger cars and tested at the laboratory experimental rig. Carrying out the study at the engine dynamometer test bench enables to reduce as much as possible the number of factors influencing measurement results and, therefore, improve the accuracy of CC evaluation. All the studied CC systems were of the same model. They were dismantled from the in-use vehicles of the same make and year of production - 2003. These vehicles were equipped with the same 1.6L 4 cylinder 4-stroke multipoint injection SI engine. No limitations have been put on vehicle's driving pattern, annual mileage, driving style etc. Only vehicles that are maintained in the authorized garage facilities were considered. Fifteen in-use cars and one new CC took a part in the study, with a mileage range from 0 to 149000 km as shown in Table 1.

The 15 CC systems dismantled from the in-use vehicles were compared with a new CC (designated number 0 hereinafter). All those 16 CC systems were mounted and tested one after the other on the test bench as shown in Figure 1. The test bench is composed of the Hoffman eddy current engine dynamometer and the Renault single point injection, 4 cylinders, 1.4L SI engine with a maximal power of 60 kW at 6000 rpm. As shown in Figure 1, the exhaust system included dual brick CC and allowed the sampling of the exhaust gases from the three different points I, II, III. The 1st sampling point was at the exit of the engine, before the first CC. The 2nd sampling point was after the first CC. The 3rd sampling point was after the whole dual brick CC system.

Table 1. In-use vehicles served as a source of the tested catalytic converters.

No	Registration date	Make of the CC	Mileage Km
0	New	Genuine, OEM	0
1	30.05.03	Genuine, OEM	62011
2	27.05.03	Genuine, OEM	62299
3	26.06.03	Genuine, OEM	63602
4	06.02.04	Genuine, OEM	65012
5	19.11.02	Genuine, OEM	80694
6	29.10.03	Genuine, OEM	84241
7	24.03.04	Genuine, OEM	86251
8	20.07.03	Genuine, OEM	91712
9	19.06.03	Genuine, OEM	99212
10	26.08.03	Genuine, OEM	100323
11	28.05.02	Genuine, OEM	104401
12	11.12.03	Genuine, OEM	106326
13	24.09.03	Non-genuine	108250
14	22.03.04	Genuine, OEM	108518
15	14.08.02	Genuine, OEM	148750

Measurements of CO, HC, CO_2, O_2 concentrations and λ values have been performed by the Bosch non-dispersive infrared gas analyzer type BEA. Concentrations of NO_x in exhaust gases were measured by the Teledyne type 200 EHTEM chemiluminescent gas analyzer. To avoid engine speed fluctuations and assure the possibility of engine operation under a steady-state regime, the electric motor of the radiator's cooling fan was powered from an external source and not from the engines alternator.

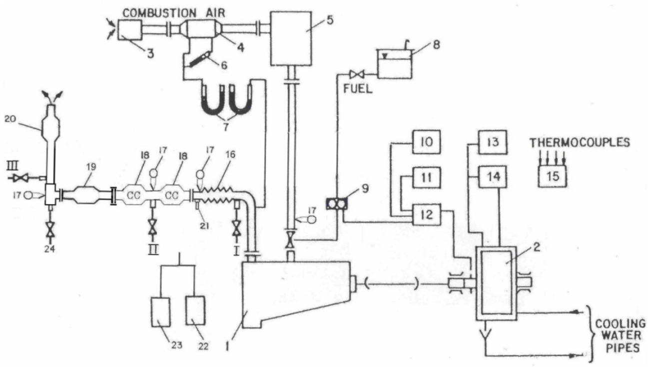

Figure 1. Layout of the engine dynamometer test bench.

1 - Engine; 2 - Dynamometer; 3 - Air filter; 4 - Air flowmeter; 5 - Plenum chamber; 6 - Inclinated manometer; 7 - U-manometers; 8 - Fuel tank; 9 - Fuel flowmeter; 10 - Engine speed indicator; 11 - Fuel counter; 12 - Dual timer; 13 - Load indicator; 14 - Servo control module; 15 - Temperature indicator; 16 - Flexible tube; 17 - Thermocouples; 18 - Catalytic converters; 19 - Resonator; 20 - Muffler; 21 - Lambda sensor; 22,23 - Gas analyzers; 24 - Water purge; I, II, III Exhaust gas sampling points.

Temperature of the engine's exhaust gas at the sampling points I, II and III, (Figure. 1), back-pressure in the exhaust system and the specific fuel consumption were measured in all experiments, as well. All measurements were performed at steady-state engine operation conditions.

The following four engine test regimes were chosen to measure the CE of in-use CC systems: 1st - power 8 kW @ 2000 rpm, 2nd - power 17 kW @ 3000 rpm, 3rd - high idle @ 2000 rpm and the 4th - low idle @ 950 rpm.

CE of CC was calculated by using the following expression:

$$E_i = \frac{(C_i^{before} - C_i^{after})}{C_i^{before}} \cdot 100\%$$

Where: E_i - CE of CC for the pollutant i; C_i^{before} - concentration of pollutant i before the entrance of exhaust gas to CC; C_i^{after} - concentration of pollutant i at the exit of exhaust gas from CC. For a dual-brick CC system three different values of the CE may be calculated for the pollutants CO, HC and NO_x as follows: E1 is the efficiency of the first CC, E2 is the efficiency of the second CC and ET is the total efficiency of the whole CC system.

The fuel used in the experiments was a commercial gasoline 95 RON, with a sulfur content of 50ppm and a density of 0.738 g/cm^3 at standard conditions. It should be mentioned that during the period between the vehicles' registration data until December 31 2004 the allowed content of sulfur in gasoline was 150ppm. From January 1 2005 until disassembling the CC systems from the vehicles, this limit was reduced to 50ppm. The allowed content of phosphorus in the Israeli engine oils has been decreased as well from 0.14% in the year 2003 to 0.08% in the year 2008.

TEST RESULTS

Based on the measurements of CO, HC and NO_x concentrations in exhaust gases, the appropriate CC conversion efficiencies were calculated for all of the considered pollutants at all engine test regimes. Figures 2,3,4 show the results of CC conversion efficiency assessment for CO, HC and NO_x, respectively. Backpressure values, as were measured at the point I (Figure 1), are given in Figure 5. Lubricant and average exhaust gas temperatures at the three sampling points I, II and III (Figure 1) are given in Table 2. Mean values of the engine-out pollutant concentrations, as were measured at all test regimes, are shown in Table 3. The mean values of the total CC conversion efficiency ET for the above-mentioned air pollutants at each one of the four test regimes were calculated. Using the Student t-distribution ($n=15$, $p<0.05$ level of significance) the "confidence interval for means" was calculated for the two loaded regimes. The results are shown in Table 4. The temperatures of the engine's lubricant at the four test regimes (Table 2) show that all the

CC systems have been tested under the same temperature conditions.

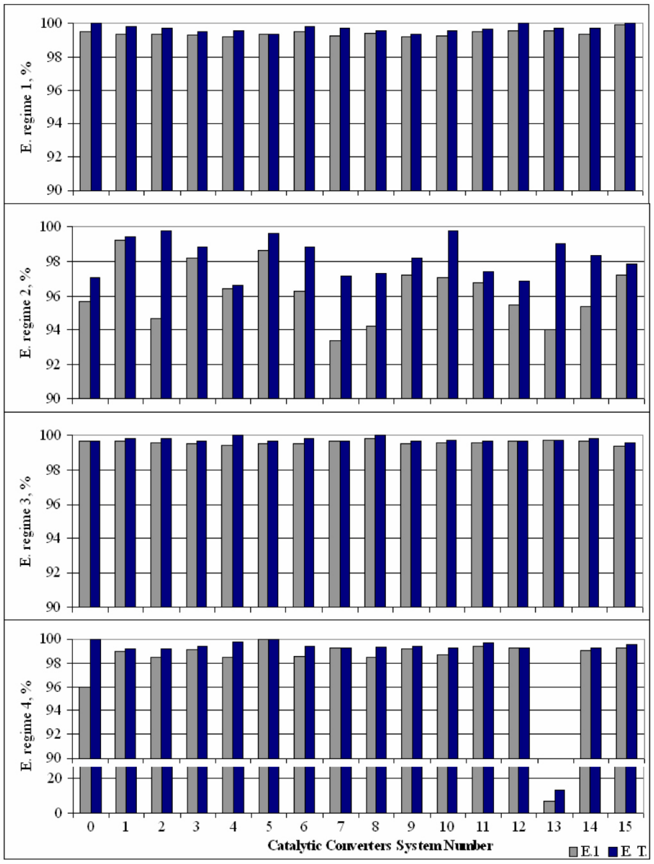

Figure 2. Conversion efficiency of CO at various test regimes.

Table 2. Lubricant and average exhaust gas temperatures.

Engine test regime	Lubricant temperature range °C	Average temperature of exhaust gas, °C		
		Point I	Point II	Point III
1	102-105	508	516	437
2	116-118	667	667	608
3	98-102	494	471	363
4	82-85	301	303	213

Table 3. Average values of the engine-out pollutant concentrations.

Pollutant	Engine test regime			
	1	2	3	4
CO, %	0.63	0.53	0.66	0.40
HC, ppm	145	110	55	208
NO_x, ppm	1750	3550	175	78

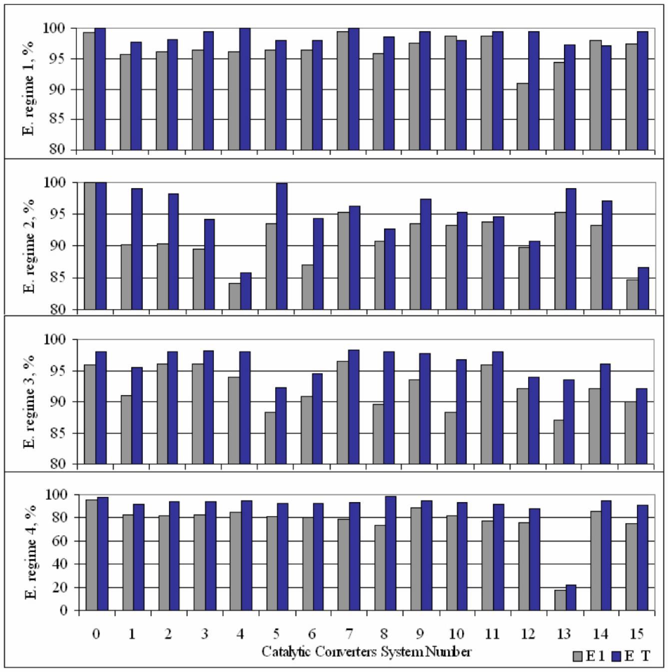

Figure 3. Conversion efficiency of HC at various test regimes.

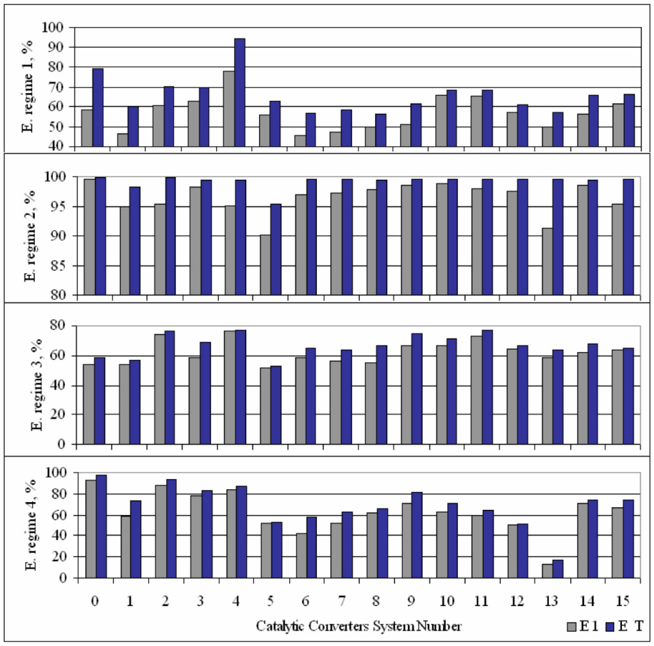

Figure 4. Conversion efficiency of NO_x at various test regimes.

Table 4. Mean values of the conversion efficiency ET, for load regimes, of 15* tested CC systems

Engine test regime	Mean Value of the Conversion Efficiency ± confidence interval, %		
	CO	HC	NO_x
1	99.36± 0.21	97.84±0.43	62.82± 4.44
2	96.60± 0.50	99.91±1.99*	95.34± 0.52

* Only a maximal efficiency value of 100% can be reached. The results are for Student t-distribution (n=15, without CC No. 13, a 95% confidence interval, P<0.05).

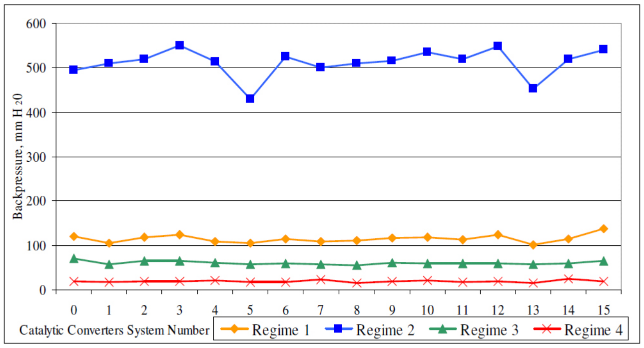

Figure 5. Backpressure as measured at the entrance to the catalytic converter system (point I).

DISCUSSION

As can be seen from Figures 2, 3, at the considered test regimes the oxidation of CO and HC occurs mainly in the first CC and only insignificantly continues in the second CC. Therefore, it may be stated that the observed values of CE for CO & HC in the first CC are, generally, similar to the CE values of the whole CC system. This similarity was observed for the testing regimes 1, 3 and 4. Only at the regime No.2 with higher engine load and speed, small quantities of CO and HC were observed as being converted in the second CC. It is interesting to note that the CC system No.13 was the only one that included an aftermarket CC. This system was also the only one, where significant catalyst deactivation was observed (the obtained results were appropriately double checked and a possibility of experimental mistake was excluded). Taking into account that CC No.13 was the only one non-genuine catalyst, it was considered an outliner and removed from the further data processing.

Concerning the total CE of CO, as shown in Figure 2 and Table 4, values of over 98% (P<0.05) were registered for the engine's test regimes 1, 3 and 4. For the regime No.2 the total CE of CO was in the range of 96% (P<0.05). The efficiency of CO conversion in the first CC has decreased at this testing regime to values lower then 93% (P<0.05). Obtained values of the total HC CE (Figure 3, Table 4) lie, in general, in the range of 89 to 99% (P<0.05) for the all test regimes. Only for the CC 4 and 15 (mileage of 65012 and 148750 km, respectively) this value decreases to 86% for the test regime No.2 (higher engine load and speed). The results of experiments show that no significant deterioration of the CC oxidation efficiency was observed for the studied mileage range until 150 thousand km.

Analysis of results presented in Figure 4, shows that NO_x conversion takes place in both CCs of the considered dual brick system. CE of NO_x has relatively low values for the test regimes 1, 3, 4, when the temperature of the exhaust gases is not high enough (Table 2). For these engine operation modes the mean CE values are in the range of 52 to 62% for the first CC and in the range of 63% (P<0.05) for the whole CC system. For the regimes 3 and 4 (idling), the minimal observed total CE drops down to 52% (excluding CC No. 13). Quite low observed conversion rates of the studied CC systems at test regimes 1, 3 and 4 can be explained by low or no engine load at these operation modes resulted in lower combustion temperatures, less NOx formation and weaker conversion process. Average exhaust gas temperatures shown in Table 2 give some indication for this explanation. For the test regime No.2 with higher engine load and speed, when the temperature of exhaust gases reaches higher values (Table 2), the CE of NOx in the first CC is over 90% for all considered CC systems. The mean total CE at this regime was found to be in vicinity of 95% (P<0.05) for the studied CC systems. The latter findings make possible the conclusion that at higher load engine operation regimes, where NOx production is high and should be treated, the considered in-use CC systems (in the studied mileage range until 150 thousand km) show quite acceptable CE without any sensible deterioration in CE as function of vehicle's mileage.

For CC system No. 13, at the test regime No.4 (low idle) CE of all three harmful components was found to be in vicinity of 20% only. The fact that only one non-genuine CC was studied, does not allow any conclusion about deterioration rate of non-genuine CC and their comparison with original ones. Carrying out this study may be an interesting continuation of the reported work.

It has to be mentioned that the observed values of CE for the CC system No. 15, with the greatest mileage of almost 150 thousand km, were quite high, as it is shown in Table 5. Values of 97-99% were reached for ET of CO; 86-99% -of HC and 64-99% - of NO_x.

Table 5. Total conversion efficiency values for the CC System No.15

Engine test regime	ET, %		
	CO	HC	NO_x
1	99	99	66
2	97	86	99
3	99	92	75
4	99	90	64

Data in Figure 5 show that the tested in-use CC systems have no sign of clogging in the studied mileage range of 150000 km.

CONCLUSIONS

1. Dependence of the catalytic converters conversion efficiency in real world usage conditions on vehicle mileage was studied for the first time.

2. It was found that the oxidation of CO and HC at all four tested regimes took place almost totally in the first CC. NO_X conversion takes place in both CCs of the considered dual brick system.

3. Conversion efficiency of NOx has relatively low mean values (52-63%) for the test regimes 1, 3, 4, when the temperature of the exhaust gases is not high enough. For the test regime 2 with higher engine load, speed and significant NOx formation, when the temperature of exhaust gases is much higher, the total CE of NOx is in vicinity of 95% for all considered CC systems.

4. CE of the CC system that has accumulated the highest mileage, of almost 150000 km, reaches values of 97-99% for CO, 86-99% for HC and 64-99% for NOx for the loaded regimes, the smaller values beeing for the low load regime when the CCs temperature is reduced.

5. No significant deterioration of the CC oxidation efficiency was observed for the studied mileage range until 150 thousand km. At higher load engine operation regimes, where NO_X production is high and should be treated, the considered in-use CC systems show quite acceptable CE without any sensible deterioration in CE as function of vehicle's mileage. Considering that the European legislation imposes a durability of 160000 km for the CCs, the results obtained in this work show that for a mileage of 150000 km the CC keeps a very high CE rate.

6. The values of the backpressure that have been registered do not show any of the CC systems as being clogged.

7. Further study of the deterioration rate of non-genuine CC and their comparison with original ones may be an interesting continuation of the reported work.

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ACKNOWLEDGMENTS

The authors gratefully acknowledge the financial support to this work of the Israeli Ministry of Environment Protection. Valuable help and cooperation provided by the Service Department of Union Motors Ltd in the supply and delivery of CC for this study is highly appreciated.

DEFINITIONS/ABBREVIATIONS

CC - Catalytic Converter

CE - Conversion Efficiency

CO - Carbon Monoxide

EUDC - Extra-Urban Driving Cycle

HC - Hydrocarbons

LEV - Low Emission Vehicle

NO_x - Nitrogen Oxides

OEM - Original Equipment Manufacturer

RON - Research Octane Number

SI - Spark Ignition

TWC - Three Way Catalyst

ULEV - Ultra Low Emission Vehicle

ZDP - Zinc-Dialkyl-Dithiophosphate