Porous Alumina-Zeolite as a Hydrocarbon Trap in SI Engine

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Abstract

Zeolite has received captivating attention as an environmental catalyst due to its capability in filtration, refining and separation of chemical species. Porous alumina structure of interconnecting channels provides the advantages by promoting radial mixing and enhancing turbulence. Existing PGM (Platinum Group Metal) catalysts, which are commonly used in honeycomb structure of conventional catalytic converters, are highly expensive. This project develops a Porous Alumina-Zeolite (PAZ) filter and investigates its influence on hydrocarbon (HC) emission from a spark ignition engine. This filter employs polymeric sponge method to obtain porous structure with a composition of 90 vol. % alumina and 10 vol. % of zeolite. The filter is tested in the exhaust system of 1300 cc engine at different speeds from 1400 rpm to 3200 rpm. The emission is measured and compared with conventional catalytic converters. Further experiment is conducted by installing both commercial catalytic converter and PAZ filter and in their reverse position. Analysis shows reduction of emission level up to 20 % is obtained between 1400 rpm to 2200 rpm for PAZ and a combined system of both PAZ and catalytic converter. The results exhibits the potential of PAZ filters as a HC trap especially in a cold start region in which the catalyst in catalytic converter has not been fully activated yet.

Keywords: Porous Alumina, Zeolite, Catalytic Converter.

1. Introduction

Incessant increase of vehicles all over the world has contributed to the serious environmental issues due to the emitted exhaust gases from the engines. Most countries have adopted continuous stringent emission regulations and enforcing the use of catalytic converter. Since then, exhaust after treatment technology has evolved and becomes one of the effective methods in pollutant abatement. In the catalytic converter, platinum, palladium and rhodium have been widely used as catalysts in lowering the activation energy of chemical reaction without changing its characteristics. However, it does not work during the first 1-2 minutes after the start of the engine due to the low temperature condition. This cold start emission is identified to contribute 60-80 % of total emission due to the inability of the present Precious Group Metal (PGM) to act below its light-off temperature (170°C for fresh catalyst and 200-225°C for aged catalyst) (1). Strategies have been adopted to enhance the capability to reduce the noxious gases. One of the effective measures are the introduction of hydrocarbon trap precedes the three-way catalytic converter in the exhaust system.

Typical system of HC trap system consists of a small volume “start catalyst”, HC adsorber and main catalyst (2). It comprises of honeycomb monolith with square channels. At the beginning when the temperature is low, HC adsorber traps hydrocarbons from the exhaust gas before they reach the main catalyst. This system requires the main catalyst to reach the light-off temperature before desorption from the HC adsorber occurs as the temperature reaches around 200°C. At the same time, as much as possible the exhaust gas should have reached HC adsorber to enable the effective reduction of emission. However, this system is difficult to optimize due to the present of HC adsorber reducing the main catalyst heating up. Another system, hybrid absorbers have been proposed as it combines both oxidation catalyst and HC adsorber in a single catalyst (2).

In HC adsorber, zeolites have been used due to its ability to selectively absorb molecules based primarily on a size exclusion process. It is commonly used in water purification, molecule separation, catalysts and specific gas separation. It exists naturally as a mineral and has been synthesized into more than 150 types of zeolite (3). The effectiveness of zeolite as molecular sieves has been investigated widely (4)(5)(6) on different particular system. P-zeolite, ZSM-5, Y-zeolite and β-zeolite represent synthetic types while mordenites, chabazites, faujasites and clinoptilolites are natural products with vast potentials as molecular sievers in HC adsorber (7).

Commercial catalytic converter employs honeycomb structure, diameter 1~2 mm which turn the flow into laminar region. Research on structure of substrate received considerable attention in order to make the flow turbulent (8). Turbulence is preferred as it improves chemical reaction. To enhance turbulence, porous structure has the potential to be employed in the catalyst design. Its open cell with interconnected channels offers low-pressure drop, radial mixing and tortuous flow paths which enhance the turbulence flow (9).

This paper reports the investigation of a hydrocarbon trap (hybrid type) in using zeolite as an HC adsorber in the porous structure to treat exhaust gases from gasoline engine. Pressure drop analysis has been performed earlier before the measurement of HC emission in engine testing. Comparison has been made to the existing catalytic converter. The effectiveness of the system is also compared to the combination of this filter and catalytic converter.

2. Experimental Work

2.1 Preparation of PAZ

The PAZ filter was fabricated using a polymeric sponge method. 90% of commercial alumina and zeolite of clinoptilolite type were the raw materials for slurry preparation. Ovalbumin with 10% of water was used as carrier and binder for the ceramic slurry. The ceramic slurry was stirred until a uniform mixture obtained. Additional binder was also added into the slurry composition. The next step was followed by the impregnation of polyurethane sponge with the ceramic slurry. The sponge was compressed to remove air and then immersed into the slurry and allowed to expand. This process was repeated to achieve the required ceramic loading. Excess slurry needs to be removed from the sponge. After that, the sponges were dried in microwave oven before went through sintering process at 1350°C. Figure 1 depicts the filter after the sintering process. Ten pieces of filters were placed in the casing for both pressure drop and emission test.

Fig. 1: PAZ filter

2.2 Pressure drop analysis

A test rig was built as indicated in Fig. 2 to perform the pressure drop measurement of the filter. Consists of a blower with a 25 hp motor, it was connected to a test section with the filter installed in its middle section. Air velocity was measured using a pitot-static probe at the inlet pipe before the test section. The measurement was begun with the velocity of 12.7 m/s until 43.2 m/s with 2.5 m/s interval. Pressure loss due to the filter was determined by the difference in static pressure obtained.

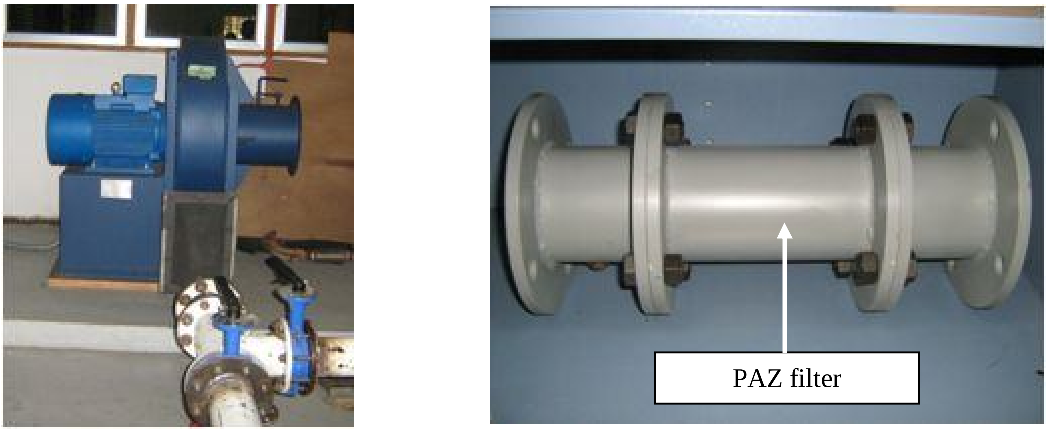


Fig. 2: A blower (left) and a test section containing a PAZ filter (right)

Pressure drop of honeycomb monolith was calculated based on the correlation obtained by Mikino et. al [11]. The given correlation of pressure drop, ΔP (mm.H_2O) is

$$\Delta P = 5.224 \times 10^{-2} \times \frac{L^{0.829}}{H.D.^{1.631}} \times \left(\frac{V}{O.F.A.}\right)^{1.405}$$

V : Gas Velocity (m/s)
Q : Gas Flow Rate (N/min^3)
L : Substrate Length (mm)
D : Substrate Diameter (mm)
HD : Hydraulic Diameter (mm)
OFA : Open Frontal Area

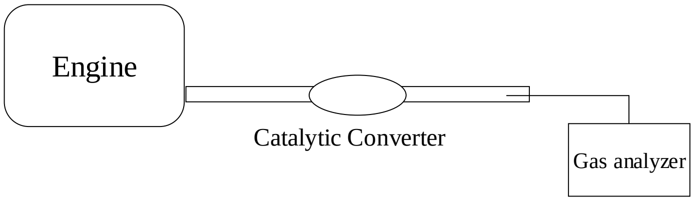
2.3 Emission Test

The emission testing was conducted on the gasoline engine (model 4G13), 1300 cc, 4-cylinder, 4-stroke and water cooled. Each exhaust system was connected directly to the engine. Techno Test gas emission analyzer was used to measure the emission from the engine. A catalytic converter was used as a reference test was a honeycomb ceramic type with 400 cells per square inch (cpsi) and 12 mils (0.012 inch) of wall thickness (400/12). The filter was installed into the casing as in Fig. 3.

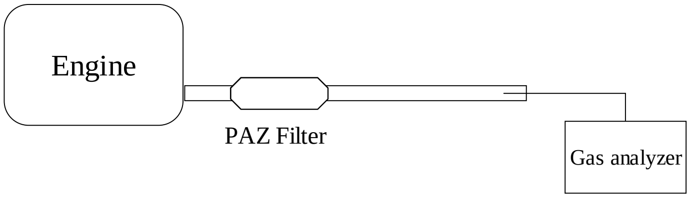
In configuration 1 of Fig. 4, considering the no load condition, the experiment was conducted by varying the engine speed. Initial speed was set to 1400 rpm with the increment of 200 rpm. At each speed, HC emission was recorded until the maximum speed at 3200 rpm. The test was then continued by changing the catalytic converter with PAZ filter as in configuration 2, followed by different setup as in configuration 3 and 4.

Fig. 3: PAZ filter installed inside the casing

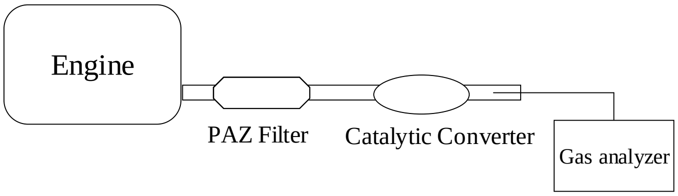
Four configurations of the exhaust system were shown up in Fig. 4.



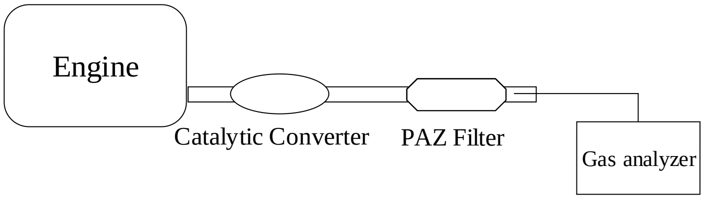
Configuration 1: Catalytic converter in the exhaust system.



Configuration 2: PAZ filter in the exhaust system.



Configuration 3: PAZ filter precedes the catalytic converter in the exhaust system.



Configuration 4: Catalytic converter precedes PAZ filter in the exhaust system.

Fig. 4: Four configurations in the emission testing of PAZ filter

3. Results and Discussion

Two essential criteria in the development of filter in automotive application is the pressure loss and conversion efficiency. Pressure loss is definitely critical as high back pressure will reduce the engine performance. On the other hand, conversion efficiency is related to the surface area for reaction to occur. Higher surface area will definitely increases the pressure loss. Therefore, both criteria need to be balanced in order to fulfill its function as filtration exhaust gas without affecting engine performance.

Fig. 5 indicates the pressure drop of the PAZ filter obtained in the experiment with the increment of air velocity. Comparison is made to the correlation obtained by previous work [11]. Three honeycomb different cell density is calculated to obtain pressure drop for 400/12, 500/12 and 600/12. It is lucidly clear that an increase of cell density raises the pressure drop of the substrate. Comparison to the experimental values of PAZ filter indicates the pressure loss is acceptable as it lies between 400/12 to 600/12 which has been widely used in the market. As a result, PAZ filter is found to be acceptable for further testing on the engine.

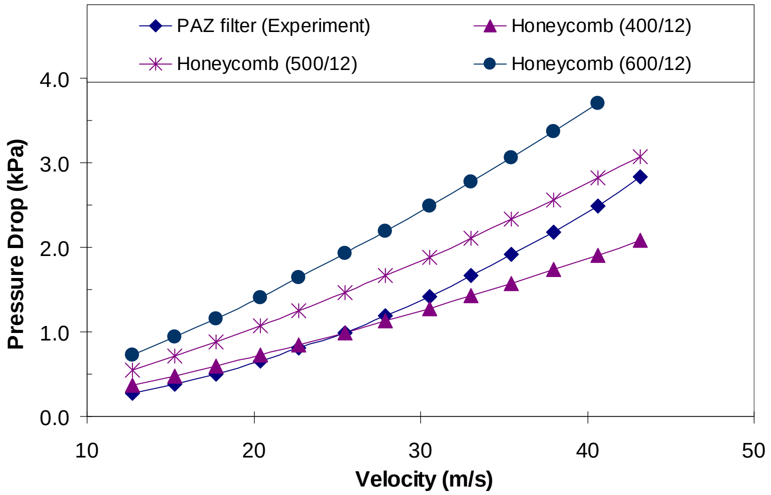


Fig. 5: Comparison of pressure drop between PAZ filter and honeycomb monolith with different cell density

In term of surface area, PAZ filter has the advantage of having interconnecting channels provides higher surface area compared to honeycomb. In this work, the effectiveness of three different configurations are compared to commercial catalytic converter. The results are plotted as normalized emission by dividing HC concentration of the tested configuration to the HC concentration of catalytic converter (if the value is larger than 1.0, then the emission is higher than the emission from catalytic converter alone. If the value is less than 1.0, then the emission is better than catalytic converter).

In Fig. 6, all three configurations show better HC emission between 1400 to 2200 rpm speed of engine. However, beyond 2200 rpm the HC filtration is no longer relatively effective except for PAZ filter. As a result, better emission reflects the potential capability of PAZ filter in trapping HC at the early stage after the engine starts.

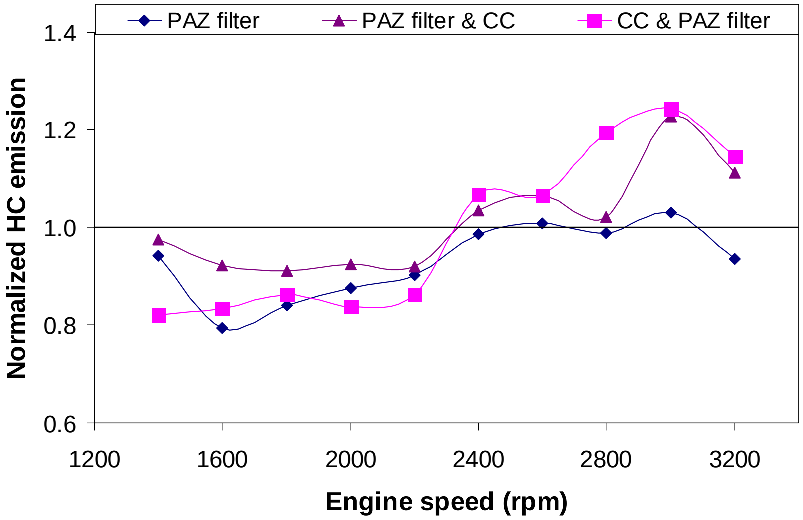


Fig. 6 HC normalized emission vs. engine speed for different configurations

4. Conclusion

Both experiments signify the potential of PAZ filter in the gasoline engine application. The pressure drop is acceptable as it ranging between 400/12 and 600/12 of commercial catalytic converter. The emission testing conducted on the engine clarify that the independent installation of PAZ filter gives lowest emission compared to other combinations. Therefore, hybrid system is preferred as the function of HC adsorber and main catalyst is combined into a single substrate.

5. Acknowledgement

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6. References

(1) Patent No.: US 6,617,276 B1, Hydrocarbon Trap/Catalyst for Reducing Cold-Start Emissions from Internal Combustion Engines.

(2) Koltsakis, G.C. and Stamatelos, A.M., Modeling of hydrocarbon trap systems, *Society of Automotive Engineers*, (2000-01-0655), CD-ROM.

(3) Weitkamp, J. and puppe (Eds.), L., Catalysis and zeolites : fundamentals and applications, (1999), p.327-329, Springer.

(4) Murakami, K., Tominaga, S., Hamada, I., Nagayama, T., Kijima, Y., Katougi, K. and Nakagawa, S., Development of a high performance catalyzed hydrocarbon trap using Ag-Zeolite, *Society of Automotive Engineers*, (2004-01-1275), CD-ROM.

(5) Nakagawa, S., Minowa, T., Katogi, K., Higashiyama, K. and Nagano, M., A new catalyzed hydrocarbon trap control system for ULEV/SULEV standard, *Society of Automotive Engineers*, (2003-01-0567), CD-ROM.

(6) Ballinger, T.H. and Andersen, P.J., Vehicle comparison of advanced three-way catalysts and hydrocarbon trap catalysts, *Society of Automotive Engineers*, (2002-02-0730), CD-ROM.

(7) Patent No.: US2005/0166581 A1, Molecular sieves for improved hydrocarbon traps.

(8) Hirose, S., Yamamoto, Y., Miyairi, Y., Makino, M., Nakasuji, Y. and Miwa, S., Application of converter efficiency simulation tool for substrate design, *Society of Automotive Engineers*, (2004-01-1487), CD-ROM.

(9) Patcas, F.C., Garrido, G.I. and Czarnetzki, B.K., CO oxidation over structured carriers, *Chemical Engineeering Science 62*, (2007), pp. 3984-3990.

(10) Makino, M. and Kondo, T., Automotive emission control technology, AVECC 2001 Asian vehicle emission control conference, 2001.