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# Selective Catalytic Reduction of Nitric Oxide in Diesel Engine Exhaust over Monolithic Catalysts Washcoated with Bimetallic Cu-Zn/ZSM-5

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## Abstract

Selective catalytic reduction (SCR) of nitric oxide (NO) in diesel engine exhaust over Cu-Zn/ZSM-5 washcoated ceramic monolithic catalysts is reported. The washcoat component was prepared by ion-exchanging ZSM-5 (Si/Al=40) with zinc while copper was incorporated through impregnation. The dispersed washcoat component was then incorporated into 400 cpsi ceramic monolith through a dipping process with the final loadings between 19.6 wt. % and 31.4 wt. %. The SCR process was studied with a feed comprising of 900 ppm NO, 2,000 ppm iso butane and 3 % oxygen at gas hourly space velocities (GHSV) between 5,000 and 13,000 $h^{-1}$. NO conversion increased until a loading of 23.6 wt. % to give a conversion of 88 % at 400 °C. The activity dropped at higher loadings due to the partial blockage of cell openings and diffusion limitations while unstable washcoating adherence was also demonstrated. After an initial deactivation of about 10 % in the first 48 h, this catalyst showed stable residual activity. Between 325 and 375 °C, minimal effect on the activity was detected when the space time was reduced from 0.94 s to 0.24 s, suggesting the absence of external mass transfer limitations for up to a GHSV of 16,000 $h^{-1}$.

***Keywords:*** NO; selective catalytic reduction; ZSM-5; ceramic monolith; performance.

## 1. Introduction

Diesel engine owes its popularity to its high fuel efficiency, reliability, durability and relatively low fuel price. This engine is run under oxygen excess condition in a so called lean-burn operation. However, the operation increases the production of toxic gas e.g. nitric oxides (NO) which cause severe environmental and health problems. Catalytic processes have been widely investigated for $NO_x$ removal and selective catalytic reduction (SCR) being the most popular one (Solis *et al.*, 2001; Pisarello *et al.*, 2002; Bennici *et al.*, 2005). The SCR technology is still immature and several drawbacks are yet to be satisfactorily addressed.

For a de$NO_x$ unit, pressure drop is a key issue (Odenbrand *et al.*, 1999; Makkee *et al.*, 2002). In a car exhaust or a power plant flue gas system, the pressure drop should ideally be below 10-20 mbar (Seijger *et al.*, 2001). Additional demands comprise low sensitivity to dust and resistance to thermal shock. Structured packings, like monolithic are gaining interest for application in $NO_x$ reduction (Zamaro *et al.*, 2005; Bueno-Lopez *et al.*, 2005) as the pressure drop in these catalysts is significantly lower than that of randomly packed bed catalysts (Cybulski *et al.*, 1999). Among various types of monolith, ceramic monoliths being the most widely used substrate material, mainly because of its relatively low manufacturing costs and high thermal stability (Heck *et al.*, 2001). However, the specific surface of most structured supports is below 1 $m^2/g$, which is way too low for catalytic purposes. The specific surface area can be enhanced up to about 40 $m^2/g$ by washcoating with suitable microporous or mesoporous materials. As zeolites have a specific surface area of 300-700 $m^2/g$, a monolayer of zeolite may serve the need for surface area and porosity (Seijger *et al.*, 2001). The ZSM-5 washcoated ceramic monolith catalysts provide interesting advantage in the SCR of $NO_x$ starts receiving attention among researchers (Zamaro *et al.*, 2005; Li *et al.*, 2008). ZSM-5 possesses high surface area of about 370 $m^2/g$ and has been demonstrated to have high hydrothermal stability and suitability in many environmental catalysis applications in our earlier works (Deeng *et al.*, 2004; Abdullah *et al.*, 2003; Abdullah *et al.*, 2006).

Among metals studied as the active component of the catalyst are Cu (Deeng *et al.*, 2004), Pt (Ismail *et al.*, 2002) and Co (Ren *et al.*, 2002). By reviewing the results, Cu should provide superiority on the basis of high activity, high stability, low toxicity and low cost. Therefore, it was used in this study. Recently, the bimetallic catalysts have created particular interest due to the promoting effect between metal species (Ismail *et al.*, 2002). The second metal should be able to maintain the first metal in the active state whereby when one of the metals is easily reduced, the other stays in a low oxidation state (Guczi and Kiricsi, 1999). The bimetallic formulations generally have higher resistance towards metal sintering and loss of contact between metal and support whereby in this case. The addition of a second metal can prevent active metal migration (Guczi and Kiricsi, 1999). Furthermore, the addition of a second metal could modify metal dispersion. In this respect, Zn was reported to improve the activity of $Rh/TiO_2$ catalysts (Hesagawa *et al.*, 2005) and Co/ZSM-5 (Ren *et al.*, 2002). The effect was mainly associated with the retardation of the reductant oxidation during the SCR process. In the present study, Cu was selected on the basis of its low cost, high activity and high stability as reported in various references (Ismail *et al*., 2002; Deeng et al., 2004; Bennici *et al*., 2005; de Lucas *et al*., 2005). Meanwhile, Zn has been demonstrated to be beneficial in improving the activity and stability of Cu/ZSM-5 catalyst in our earlier series of study as reported in Abdullah (2008).

The objective of this work was mainly to develop a ceramic monolithic catalyst for the SCR of NO. It was performed by studying the optimum loading of active porous catalytic washcoat comprising of low cost bimetallic Cu-Zn in ZSM-5 as the porous component of the catalyst. The washcoat composition and preparation method was first optimized through our earlier series of study (Abdullah, 2008). The performance of the washcoated ceramic monolith catalyst over long time on stream as well as over various gas hourly space velocities (GHSV) were to be demonstrated.

## 2. Materials and Methods

### 2.1. Preparation of Cu-Zn/ZSM-5 washcoat

First, Cu/ZSM-5 was prepared by impregnating ZSM-5 (Zeolyst International, USA, Si/Al=40) using $Cu(NO_3)_2\cdot 3H_2O$ (Merck, Germany) solution. The slurry was placed in a rotary evaporator and kept under vacuum at 100 °C until complete dryness and free flowing. It was subsequently dried at 100 °C for 24 h and then calcined at 600 °C for 2 h. Then, the Cu/ZSM-5 was subjected to a second metal incorporation process whereby Zn was ion-exchanged into the zeolite using $Zn(NO_3)_2\cdot 6H_2O$ (Merck, Germany) as the precursor. The mixture was then stirred continuously at room temperature for 24 h. Then, the suspension was filtered and dried at 100 °C for 24 h, followed by calcinations at 600 °C for 2 h. The ion-exchange process was repeated until 8 wt. % of Zn was incorporated into the zeolite. The amount of aqueous solution per gram of zeolite used in this study was as reported by de Lucas *et al.* (2005) and Nejar *et al.* (2007) for the ion-exchange and impregnation process, respectively. This resulted in Cu-Zn/ZSM-5 catalyst washcoat component with 6 wt. % and 8 wt. % of Cu and Zn, respectively.

### 2.2. Preparation of washcoated monolithic catalyst

Slurry of washcoat component was prepared by wet mixing. During this process, 20 wt. % Cu-Zn/ZSM-5 washcoat component was dispersed in a solution of methyl cellulose (Acros Organics, 2 % solution in water, 4000 cP) and deionized water (1 wt. % of methyl cellulose in solution) and stirred using magnetic stirrer 6 h to inhibit solvent-particle interactions (Pfeifer *et al.,* 2005). The slurry was then applied to the 400 cell per square inch (cpsi), 0.17 mm average wall thickness and 2.0 cm diameter and 6.0 cm long ceramic monolith (Guide Ceramics, China) (shown in Fig. 1) by dip coating method using self-fabricated dipping equipment. Based on method reported by Beers *et al.* (2003), the ceramic monolith substrate was dipped in the slurry for 3 min (varying the time from 5 s to 5 min did not affect the loading) and withdrawn at a speed of 40 cm/min. Subsequently, the washcoated ceramic monolithic was partially dried

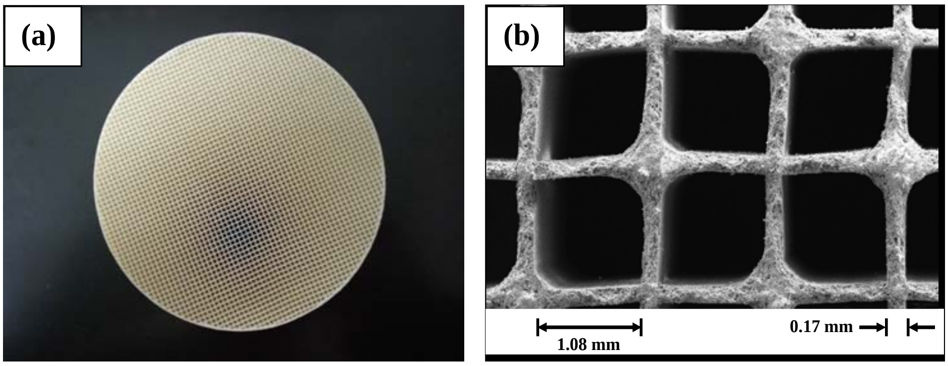

Figure 1. Picture of the ceramic monolith substrate used (a) and enlarged image of the cells (b).

Table 1. Washcoat loading and weight loss for different repeated dipping processes and weight loss after ultrasonic treatment.

| Sample | Amount Loading (g) | *Percent Loading (wt. %) | **Weight Loss (%) |
|---|---|---|---|
| 1 | 1.5 | 19.6 | 3.3 |
| 2 | 1.8 | 23.6 | 5.6 |
| 3 | 2.2 | 28.8 | 31.8 |
| 4 | 2.4 | 31.4 | 37.5 |

*Based on monolith substrate weight of 7.64 g.

**Based on amount of washcoat component loaded.

at room temperature for 1 h. The dip-coating was repeated for up to 4 times to prepare 4 different monolithic catalysts with loadings between 19.6 and 31.4 wt. % which was measured after calcinations in a furnace at 450 °C for 5 h. The catalyst loading was determined by measuring the weight increase of the ceramic monolith before and after the washcoating process.

### 2.3. Washcoating adherence test

The adherence of catalyst coatings was evaluated using a method as described by Boix *et al.* (2003) and Zamaro *et al.* (2005). It was based on the measurement the weight loss that resulted after exposing the sample to ultrasound treatment. In this work, the coated monoliths were subjected to ultrasound treatment by immersing them in acetone inside a glass vessel and then placed in an ultrasonic bath for 1 h at 25 °C. After that, the samples were rinsed and oven-dried at 120 °C for 2 h before weighing.

### 2.4. Catalyst activity measurements

The SCR experiments were carried out in a continuous flow reactor system. This set-up consisted of a series of feed gases with individual control by mass flow controllers, a catalytic reactor heated with a tubular furnace and an exit gas flow meter equipped with gas analysis system. The monolithic catalysts were brought to reaction conditions for each test (900 ppm NO, 2000 ppm iso-butane, 3 % oxygen and the balance $N_2$, a space velocity (GHSV) of 13,000 $h^{-1}$ and a total flow rate of 150 ml/min). This gas composition was actually a simulated composition of typical diesel engine exhaust gas. The temperature of the catalyst bed was taken as the reaction temperature which was measured by means of a K-type thermocouple. The feed gas mixture was allowed to flow in to the reactor containing the monolithic catalyst for about 30 min under desired the reaction conditions and the concentration of gases at the inlet and outlet was determined using gas chromatography (Shimadzu GC 8A) and gas analyzer (KANE-MAY 900, England). A series of experiment were carried out at either various reaction temperatures (300 °C to 400 °C) or gas hourly space velocities (GHSV). The space velocity was varied by changing the total feed gas flow rate from 50 to 200 ml/min with a corresponding GHSV of between 4,000 $h^{-1}$ to 16,000 $h^{-1}$. This flow conditions led to a corresponding space time of 0.24 s to 0.94 s.

## 3. Results and Discussion

### 3.1. Washcoating adherence

Table 1 shows the increasing washcoat loading into the ceramic monolith substrate upon repeated dipping process. The first dipping process resulted in 19.6 wt. % loading and an increase by merely 4 wt. % resulted in the subsequent dipping process. Subsequent dipping steps resulted in monolithic catalysts with up to 31.4 wt. % of washcoat layer. Significant loading of the washcoat after the first dipping was attributed to its significant adherence to wall surface the monolith substrate. The dry condition of the monolith resulted in the rapid adsorption of moisture (from the washcoating slurry) into the pores of the wall, leading to good adherence. Generally, coating was thinner and more evenly distributed as shown in Fig. 2. Subsequent dipping would coat on the first washcoat layer that was not so well-distributed and had lower surface area. Consequently, only an increase of about 3-4 wt. % resulted in every subsequent dipping steps. It should be noted that during the second to the fourth dipping step, the adherence of the washcoat layer primarily occurred between the new and existing outer wascoat layer while it was between the monolith and the washcoat layer in the case of the first dipping step. The former mode of adherence was deemed stronger than the latter mode of adherence.

The weight loss (%) caused by exposing the sample to ultrasound treatment to test the coating adherence is also shown in Table 1. As noted earlier, the multiple dipping processes increased the catalyst

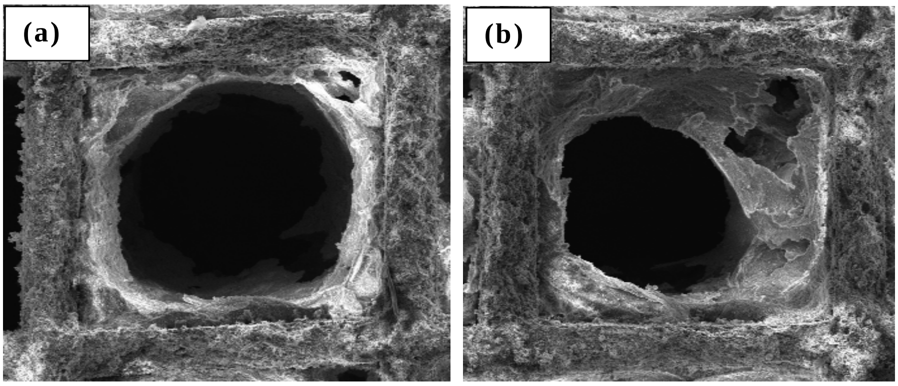

Figure 2. Typical images of the monolith cells when loaded with (a) 19.6 wt. % and, (b) 31.4 wt. % of Cu-Zn/ZSM-5.

loading in ceramic monolithic catalyst was loaded onto the monolith. However, it also led to a decrease in the stability of the washcoat as suggested by the increase in weight loss after the ultrasonic test. Significant detachment of the washcoat layer was observed when the dipping step was repeated three times (Sample 3) with a loss of about 31.8 %. Similar behavior was also demonstrated by Sample 4. This was indeed a drastic increase as compared to the loss experienced by Sample 1 and 2. In these two samples, the strong adherence between the monolith wall surface and the washcoat layer was responsible for the low weight loss. Valentini *et al.* (2001) and Boix *et al.* (2003) suggested that a weight loss of between 0 and 10 % should be considered acceptable to indicate a stable coating adherence.

This stable coating was due to the average particle size of the slurry used for coating which was significantly smaller than the macropore size of the monolith walls. So, it was expected that when the monolith was dipped in the catalyst slurry for the first time, a large amount of catalyst was absorbed by the cordierite pores and was retained inside the monolith structure after calcinations as shown in Fig 1(a). The third and fourth immersion steps caused a significant loss of adherence of the catalyst coated onto the substrate. This was due to the thickness of the catalysts covering the surface of the ceramic monolith which increased with an increase in the number of immersion as shown in Fig 1(b). This resulted in weak adherence of the catalyst and the surface of the substrate. Thus, 1.8 g catalyst resulted from the twice dipping process is the optimal loading and gave the stable of catalyst coating based on the ultrasound treatment.

### 3.2. Catalytic Study

#### 3.2.1. Optimal loading of washcoat layer

The profiles of NO reduction versus reaction temperature, with optimum points in the 300-400 °C range are shown in Fig 3. Generally, the trend is characterized by low removal at low temperature due to low catalytic activity and also low removal at high temperature due to the combustion of iso-butane that acted as the reductant for NO in the SCR reaction. The % NO reduction was found to increase up to a loading of 23.6 wt. % to give a NO

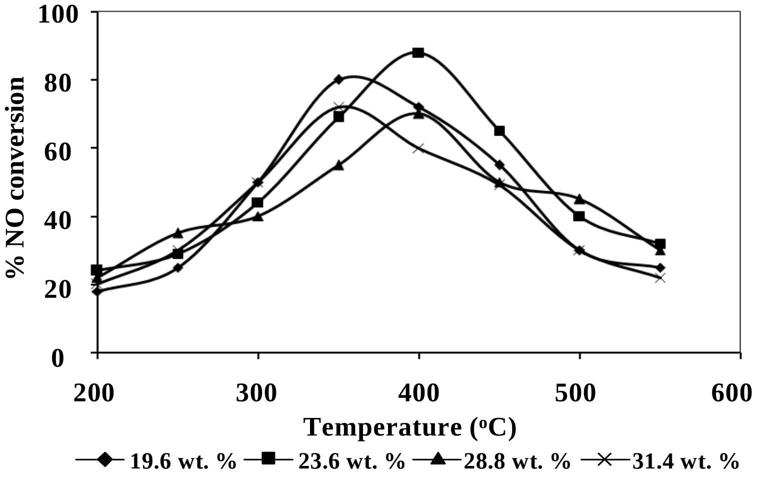

Figure 3. NO conversion profiles of monolith catalysts with various washcoat loadings. (Reaction conditions: 900 ppm NO, 2000 ppm i-$C_4H_{10}$, 3 v % $O_2$ and $N_2$ as balance).

removal of about 88 % at 400 °C while higher loadings did not result in any beneficial effect. This observation was attributed to the partial blockage of cell openings of the ceramic monolith as can be seen in Fig 2(b). The excessive accumulation of washcoat also meant that significant portion of the mass would be unavailable for contact with the feed gas.

The performance ceramic monolithic catalyst for SCR of NO used in the present study was comparable with results reported in the literatures using similar ceramic monolith but washcoated with different types of catalytic washcoat layer. Boix *et al.* (2003) reported that Pt/Co/FER-washcoated ceramic monolith merely achieved about 17 % of $NO_x$ reduction while the corresponding powder Pt/Co/FER catalyst showed a maximum $NO_x$ conversion of 45 %. Krocher *et al.* (2006) reported that Fe/ZSM-5 coated on ceramic monolith exhibited over 80% $NO_x$ reduction between 400 to 650 °C which was slightly higher than the optimum temperature obtained in this study. Although a fair comparison is difficult as the reactions were conducted under slightly different conditions, the monolithic catalyst used in present study demonstrated impressive results in NO removal from diesel exhaust. Thus, the ceramic monolith catalyst loaded with 23.6 wt. % of Cu-Zn/ZSM-5 washcoat layer was used for further investigation in terms of its stability and applicability in wider GHSV range.

### 3.2.2. Stability study of the monolith catalyst

Fig. 4 shows the stability of the catalyst over long range of time on stream. It is noted that NO conversion decreased by merely 10 % from 85 % to 75 % in the first 48 h. Deeng *et al.* (2004) reported that wiremesh structured catalyst was able to maintain its activity for NO reduction for 48 h. In the present study, the duration was extended to 72 h and the monolithic catalyst showed a stable residual NO conversion of about 73% after 48 h on stream. The initial drop in the catalytic activity was ascribed to the partial sintering of the washcoat layer leading to minimal loss of active surface area. Sintering is commonly reported to cause initial drop in the activity for SCR of $NO_x$ especially when operated above 300°C (Ren *et al.*, 2002; Hesagawa *et al.*, 2005). In the study, the monolithic catalyst was exposed to a reaction temperature of 400°C and therefore, partial sintering was unavoidable. However, the effect to the activity was very minimal. Stable activity at long time on stream demonstrated the high thermal, mechanical and chemical stabilities of the monolithic catalyst for this application. In this respect, this catalyst was deemed to be more superior than the monolithic catalyst washcoated with other types of zeolite such as beta (Beers *et al.*, 2003; Bueno-Lopez *et al.*, 2005), mordenite (de Lucas *et al.* 2005) or Ferrierite (Boix *et al.*, 2003). These zeolites often subject to drawbacks of having either low surface area, low hydrothermal stability, high coking activity due to high acidity or higher tendency to be deactivated by coke due to pore system of different sizes (Chen, 1994).

### 3.2.3. Effect of space time

Fig. 5 shows NO concentration versus space time of the feed in the reactor at different temperatures. It approached a third order polynomial curve within the temperature range tested. It is characterized by rapid drop in the NO conversion at very low space time (or very high GHSV) to suggest the reaction controlling phenomenon within this range. Beyond a space time of about 0.24 s to the maximum used in this study i.e. 0.94 s (corresponding to a GHSV of 16,000 $h^{-1}$), minimal effect was observed with decreasing flow rate (increasing space time). This was an indication of the absence of internal mass transfer limitations in the

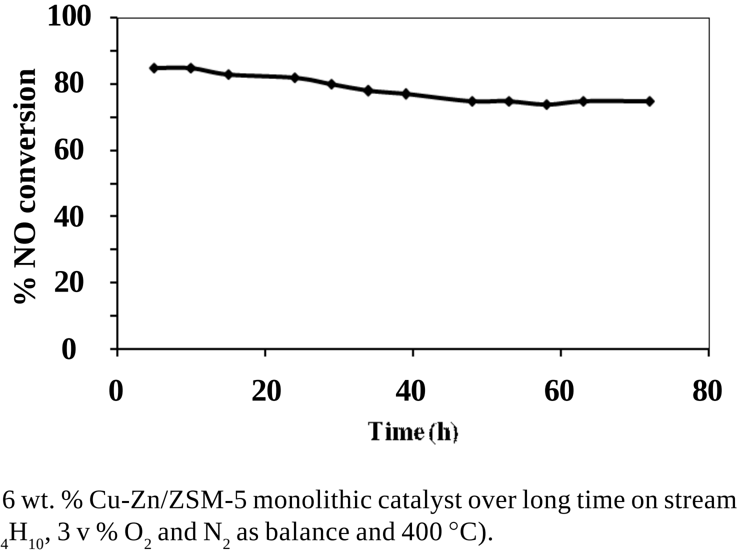

Figure 4. Stability of 23.6 wt. % Cu-Zn/ZSM-5 monolithic catalyst over long time on stream. (Reaction condition: 900 ppm NO, 2,000 ppm i-$C_4H_{10}$, 3 v % $O_2$ and $N_2$ as balance and 400 °C).

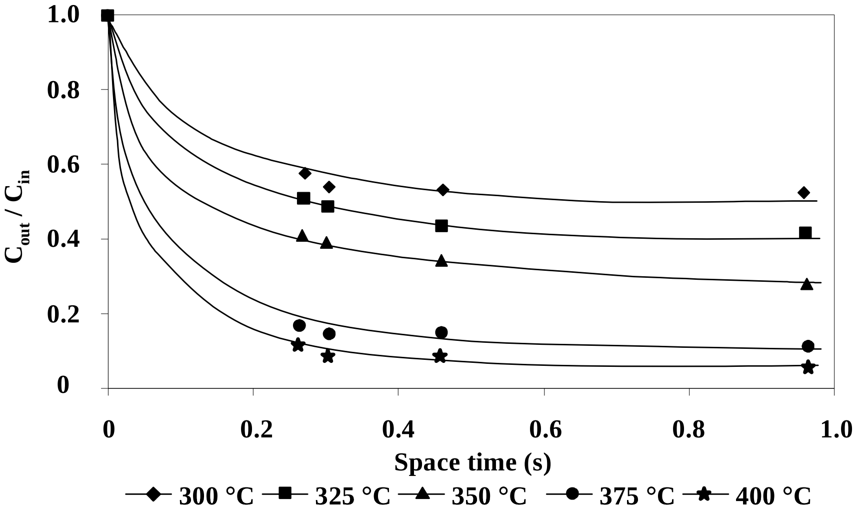

Figure 5. Dimensionless concentration of NO in the outlet as a function of space time at different reaction temperatures. (Reaction condition: 900 ppm NO, 2,000 ppm $i$-$C_4H_{10}$, 3 v % $O_2$ and $N_2$ as balance).

reaction system at high space time (Li *et al.*, 2008). This conclusion was at least valid within a temperature range of 300 to 375 °C. The presence of external mass transfer limitations within this range was unlikely as the feed composition was fixed throughout the study.

In general, the optimum monolithic catalyst washcoated with Cu-Zn/ZSM-5 used in the present study demonstrated the high catalytic activity in SCR of NO in the feed stream over a wide range of space time (or GHSV correspondingly). The absence of internal and external mass transfer limitations which are very critical in the operation of a catalytic reactor were achieved with a GHSV as high as 16,000 $h^{-1}$ to suggest the potential of this type of catalyst for NO removal process from diesel exhaust.

## 4. Conclusions

A washcoated ceramic monolith catalyst should have good adherence between the washcoat component and the monolithic substrate. The present work found that 23.6 wt. % of Cu-Zn/ZSM-5 washcoat component containing 6 wt. % of Cu and 8 wt. % of Zn washcoated onto the 400 cpsi ceramic monolith produced a monolithic catalyst with optimal performance. It maintained a good adherence to the monolithic substrate when ultrasonic treatment was applied while the catalytic performance achieve a ~88 % of NO reduction at 400 °C. The activity dropped at higher loadings due to the partial blockage of cell openings and diffusion limitations while unstable washcoating adherence was also demonstrated. After an initial deactivation of about 10 % in the first 48 h, this catalyst showed stable residual activity. Between 325 and 375 °C, minimal effect on the activity was detected when the space time was reduced from 0.94s to 0.24 s, suggesting the absence of external mass transfer limitations for up to a GHSV of 16,000 $h^{-1}$. This study successfully demonstrated the high activity and stability of monolithic catalyst washcoated with Cu-Zn/ZSM-5 for SCR of NO in diesel engine exhaust gas.

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