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CATALYTIC CONVERTER MATERIAL TREATED BY ULTRASONIC APPROACH

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ABSTRACT

Automobile catalytic converter is a component to reduce the pollutant production such as Carbon Monoxide (CO), Nitrogen Oxides (NO_x) and Hydro Carbon (HC). Material for catalytic converter is selected. However, the challenge is how to select the right materials and right methods. This research approaches the selective materials such as the γ- Al_2O_3 nanocrystalline as the wash coat, NiO as the catalyst and FeCrAl as a substrate. Those materials was selected due to their advantages. New method is approached by using a combination of ultrasonic technique provide more acceleration to the γ-Al_2O_3 powders. Washcoating process is conducted by using electroplating of Ni plate on FeCrAl substrate in sulphamate type as electrolyte medium. The potential result is the catalytic converter components provide the oxidation resistance in short term and long term oxidation resistance in high temperature operation as well as improving the homogeneity of material/wall thickness.

Keywords: catalytic converter, γ-Al_2O_3 wash coat, NiO catalyst, FeCrAl substrate, Ultrasonic technique.

1. INTRODUCTION

There are several products of the gasoline engine operation such as CO, NO_x and HC as unburned compounds. Those products lead to air pollutant that high contribute to the environment problems [1-4]. The total pollutant which is caused by transportation sector for the years 1997-2000 in Malaysia is shown in Table-1 [5].

Table-1. Pollutant value during 1997-2000 [5].

Pollutant	CO (tons)	NO_x (tons)	HC (tons)	SO_x (tons)	Particle (tons)
1997	1.9 mil	224,000	101,000	36,000	16,000
1998	2.0 mil	237,000	111,000	38,000	17,000
1999	1.9 mil	268,000	120,000	No info	17,480
2000	2.3 mil	302,678	141,000	40, 126	19,277

Exhaust gas pollution limits in Malaysia which consequently cause motivate utilization of automobile catalytic converters. Main function of the catalytic converter is to reduce the pollutant in the automobile exhaust gases [6]. It has three basic components i.e. substrate, catalyst and wash coat as shown in Figure-1 [7].

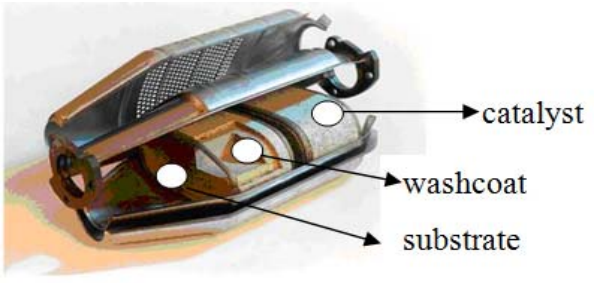

Figure-1. Catalytic converter component [3, 7, 27]

Many researchers have been explored how to develop the efficient catalytic converter [4, 6, 8-12]. Most frequently used design of the catalytic converter is a monolithic structure which is coated with a wash coat that supports the catalyst material. Several wash coat as catalyst carrier are Al_2O_3, SiO_2, TiO_2 and SiO_2-Al_2O_3 [2, 13]. There are several range of oxides which used as alternative catalyst material such as CuO, V_2O_5, NiO, MoO_3 and Cr_2O_3 because they are cheaper as compared to precious metal (Pt, Pd and Rh) [14]. In addition, most of the substrate material is FeCrAl because some advantages of its properties [3, 4, 12].

1.1. Catalytic converter material

Catalytic converter has been investigated by many researchers in order to develop the highest performance of catalytic converter to reduce the pollutants. Therefore, the exploration regarding to the materials and methods to be challenge to explored. Most of the researchers use three way catalytic (TWC) converter that was constructed by the catalyst coated pellets tightly packed in a sealed shell. The modern model vehicles are equipped with a monolith type TWC that uses a honeycomb shaped catalyst element. Less exhaust backpressure is created by monolith design and providing sample surface to convert feed gases efficiently as well

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[15]. According to Makalam *et al.,* (2009) [16] the catalytic converter were developed using catalyst material which consists of metal oxide such as TiO_2 and CoO with wire mesh substrate. Those metal oxide are inexpensive as compared to Palladium and platinum. There are two comparisons such as original engine manufacture catalytic converter (OEM catco) and wire mesh catalytic converter. The results show that the wire mesh catco reduce 24%, 41% and 40% of NO_x, Co and HC emission as compared to OEM catco, respectively. Qingbao *et al.,* (2007) [17] investigate the palladium catalyst which $Ce_xZr_{1-x}O_2$ as washcoated monoliths. The results shows that a Zr component of Pd-$Ce_xZr_{1-x}O_2$/monoliths catalyst contribute to the catalytic activity up to x=0.8>0.6>0.5≈1. It means that there are strong interaction among palladium, ceria-zirconia and the monolith. According to Silva *et al.,* (2006) [18] the temperatures of exhaust gas and substrate wall at the catalyst outlet increase with BMEP and rpm. However, Co, No_x and HC shows high conversion efficiency regardless of BMEP and rpm.

1.2. Substrate

Substrate is a support, where the catalyst is deposited. Therefore, it is frequently called as a catalyst support [19]. Main function of the substrate is to bring the maximum active catalyst [19]. It must be accommodating the rapid changes of temperature, pollutant gas, road shock and chassis vibrations. The substrate supports the catalyst which is used in exhaust of the combustion chamber which leads to obtain the low pressure drop [20]. Significant amount of substrate in accommodating the catalyst is provided by high surface area [21]. There are many substrate material used by researchers as listed in Table-2.

Table-2. Various substrate materials for catalytic converter.

No.	Substrate material	Reference	Disadvantages
1	Si, SiN, SiO_2 and Al	[22, 23]	High Defect density (10^{-6}/cm^2) and the determination of a suitable buffer layer
2	Wire mesh	[16]	The absence of radial mixing, the low interphase mass and heat-transfer rate
3	FeCrAl	[3, 4, 12, 24-26]	Crystal intergrowth in high temperature

Ferritic steel (FeCrAl) foils become the most interesting material than ceramics [3, 4, 12, 24-26]. It is selected as the substrate since the high temperature oxidation resistance and achieve larger specific surface. According to Twigg and Webster, (2006) [21] here that another function of the ferritic steel is that it can strongly adhering oxide film on their surface. The surface oxide film is developed by chromia rich when the material heated by 300-400 ^{0}C. Moreover, the alumina rich surface is developed when heated at 800 ^{0}C which is promoting the high temperature resistance.

1.3. Washcoat

There are several types of wash coat material that become interesting material for many researchers refer to Table-3.

Table-3. Various wash coat material.

No.	Washcoat material	Reference	Disadvantages
1	Pt, Rh and Pd	[1]	Sensitive to thermal degradation at high temperature
2	SiO_2, TiO_2, and SiO_2- Al_2O_3	[2, 13, 16, 25]	Low heat and mass transfer
3	γ- Al_2O_3 powders	[3, 4, 12, 25]	The deposition of γ- Al_2O_3 has not been well as compared to ceramic monolith
4	CeO_2–Mn	[25]	The trapping of sulfur which led to corrosion
5	α- Al_2O_3	[25, 28]	Segregation of an alumina layer at high temperature
6	Al_2O_3/CeO_2	[29]	Trapping of sulfur and low heat transfer

The washout is a thin layer of gamma alumina (γ-Al_2O_3) coating, typically 20-150 μm thick with a high surface area on the top of substrate and as the catalyst's carrier [30]. Several examples of a high surface area carrier are Al_2O_3, SiO_2, TiO_2, and SiO_2-Al_2O_3 [2, 13, 31]. According to Ersson and Jaras, (2006) [20] the washout is not change while extended temperature operation of the catalyst in Honeycomb Monolith substrate at the range between 1000 and 1400 ^{0}C.

1.4. Catalyst

Catalyst is material that can to improve the rate and selectivity of a chemical reaction and in the process is cyclically regenerated [33]. Sebayang *et al.* (2009) [12] explains that catalyst accelerate the chemical reaction of oxidation and reduction. The catalyst material can be summarized at the Table-4.

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Table-4. Various catalyst materials.

No.	Catalyst material	Reference	Results
1	Pt, Pd, Cu, Cr, Ni and Mn	[2, 38-40]	Enhanced metal-support interaction and keeps good dispersion after reaction as well as excellent oxidation catalysts
2	TiO_2 and CoO	[16]	Those metal oxide more inexpensive as compared to Palladium and platinum
3	nickel nitrate and nickel cetylacetonate	[28]	Increment in the dispersion of the active phase and to a more active catalyst.
4	NiO	[34]	The interactions in co-precipitated NiO-Al_2O_3 system are intensive
5	$NiWO_4$, and WO_3	[35]	In the range(523 to 723 K) NiO is more active than $NiWO_4$ and WO_3 catalysts
6	Ni/Al_2O_3	[36, 37]	Enhanced the absorption of $Ni(NO_3)_2.xH_2O$ on the surface of supporters
7	Pt and Rh	[32]	Possibly stabilized against sintering
8	Ni- and Cu/alumina	[37]	Bigger specific surface area

2. NEW METHOD APPROACHED

Concerning technique for adhering catalyst on substrate by wash coating technique is founded by some reviews [3, 4, 12, 41, 42]. Dip coating is one of the most common methods to form oxide coating thin layer on the metallic substrate. It is combined with pre-treatment such as shortened the diffusion path before depositing the wash coat. [3, 6, 43]. The other methods for preparing FeCrAl alloy supported perovskite for catalytic combustion of methane are co-precipitation, spray-pyrolysis and sol-gel methods [44]. Moreover, other technological procedure such as electrophoretic deposition [45, 46], aluminizing technique [47] and solution combustion synthesis (SCS) is basic to develop and bind to the catalyst on the FeCrAl substrate [12]. Limitations of the some methods above in powder form has become main issues in applying the catalyst. Therefore, some modifications is required. According to Sebayang *et al.,* (2009) and Y. Putrasari *et al.,* (2010) [3, 12], the methods for preparing NiO catalyst on FeCrAl substrate is by using combination of the electroplating, ultrasonic technique, ultrasonic bath and oxidation process. However, they still found the washout layer is spalling since the loose adhesion and unstable oxide growth in long term oxidation and NiO was not directly diffused to the substrate. However, NiO created through plating Ni to the substrate FeCrAl and NiO existed through oxidization in high temperature. According to Ade Firdianto, (2012) [4] the washout technique is conducted by using electroplating method. Ultrasonic technique is conducted to achieve the surface deformation and can accelerate the γ-Al_2O_3 powders. Ni electroplating is used and γ-Al_2O_3 is used as metallic washout on FeCrAl substrate in order to obtain large surface area, good pore size distribution, surface acidic properties and crystal structure. He found that the surface area on washout relatively small since the particle size in the range of 50-53 μm. In addition, the characteristic of γ-Al_2O_3 coated on the Fe-Cr-Al at low temperature between 500 700, 900 and 1100 ^{0}C is successfully conducted. However, if it applied in higher temperature operation, the transformation into α-Al_2O_3 form is inevitable. Moreover, according to Ghosh *et al*., (2008) [48] the particle size of washout decreased as surface area on wash coat increased.

All the references above have some weaknesses in each result. Therefore, this research approach the new γ-Al_2O_3 nanocrystalline as a wash coat, nickel oxide as a catalyst material and metallic FeCrAl as a substrate which is applied for catalytic converter using some combination methods. The methods of creating new wash coat of nanocrystalline γ-Al_2O_3 on FeCrAl substrate by using the combination of the ultrasonic technique between ultrasonic clamp on tubular type (Figure-2) and ultrasonic cleaning bath as shown in Figure-3 in order to obtain larger surface area, best pore size distribution and more accelerate the γ-Al_2O_3 powders as well as to reduce the possibility of γ-Al_2O_3 transformation in high temperature.

The various time of ultrasonic clamp-on process at 10, 20, 30, 60 minutes with coupled frequency of 18.52 kHz. Meanwhile the ultrasonic cleaning bath process will be conducted at the frequency of 35 kHz at the holding time of 4.5 hours.

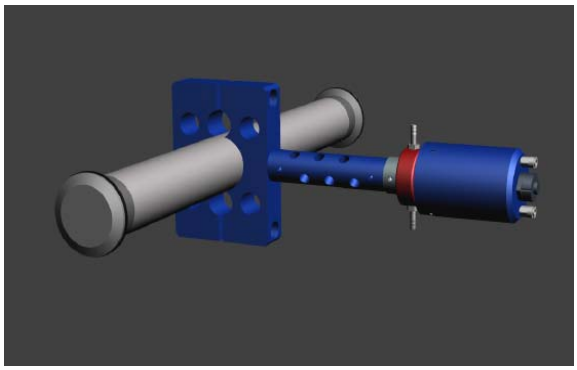

Figure-2. Ultrasonic clamp-on.

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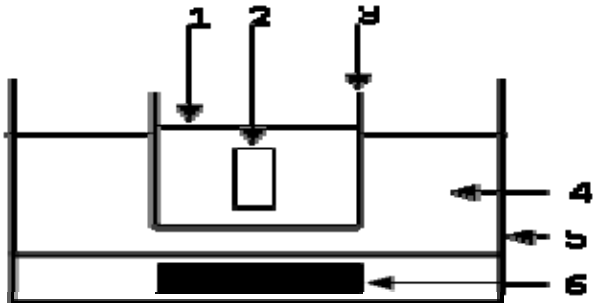

Figure-3. Schematic diagram of ultrasonic cleaning bath (1) Methanol; (2) Specimen; (3) Beaker; (4) Water; (5) Bath; (6) Ultrasonic source.

New process was conducted in creating NiO through nickel electroplating on FeCrAl substrate by using sulphamate type as electrolyte medium which is consist of nickel sulphamate ($Ni(SO_3NH_2)_2.4H_2O$), nickel chloride ($NiCl{\cdot}6H_2O$), boric acid (H_2BO_3), and sodium lauryal sulphate ($\underline{C_{12}}\underline{H_{25}}\underline{SO_4}\underline{Na}$).It is conducted at a constant temperature of 40-60 ^{0}C with PH adjusted at the range of 2.5-4.5 using HCl and NaOH reagent.

3. POTENTIAL RESEARCH

Potential of the approach research are found the new method of creating washout on new catalyst and new process for developing nickel oxide as well as washout for catalytic converter. The expected results are the γ-Al_2O_3 nanocrystalline and nickel oxide will embed on the Fe-Cr-Al metallic substrates. The new oxide of γ-Al_2O_3 nanocrystalline and NiO can able to provide oxidation resistance on coated FeCrAl in long term oxidation as well as NiO can directly diffuse into the substrate. New ultrasonic approach is used to improve the efficiency of electroplating such as in agitation process.

4. CONCLUSIONS

There are many differences of the materials and methods were explored by many researchers. Regarding to that results show their properties need to improve by using different material with new combination of ultrasonic technique. The main concern of the reviews is how to treat and choose the material for washout, catalyst and substrate in order to achieve a good oxidation resistance in long term temperature. The current washout material is γ-Al_2O_3 nanocrystalline and the NiO as the catalyst as well as FeCrAl as a substrate for catalytic converter.

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